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ULTRASONIC DETERMINATION OF  
COMBINATIONS OF THIRD-ORDER  
ELASTIC CONSTANTS OF SMALL  
CUBIC SINGLE CRYSTALS

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the material and on the cross sectional dimensions; the current estimate of the minimum length is ~ 4 mm. Diffraction corrections based on the Kirchhoff diffraction theory are presented and analyzed for both the fundamental frequency component and the second harmonic component of the ultrasonic wave. The technique was used to measure combinations of third-order elastic constants of small samples of  $\text{CsCdF}_3$  and  $\text{KZnF}_3$ . The measured combinations, in units of  $10^{11}$  dyn/cm<sup>2</sup>, obtained by correcting only the fundamental frequency component for diffraction, are as follows.

Sample	$C_{111}$	$(C_{112} + 4C_{166})$	$(6C_{144} + C_{155} + 8C_{456})$
$\text{CsCdF}_3$	-13.4	- 7.24	-46
$\text{KZnF}_3$	-16.8	-11.3	

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by


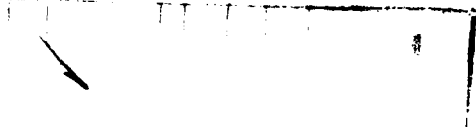
Bruce Dion Blackburn

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May 1981

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## PREFACE

This report is an adaptation of the dissertation of Bruce Lion Blackburn submitted to the Department of Physics at The University of Tennessee in partial fulfillment of the requirements of the degree of Doctor of Philosophy.

The ultrasonic harmonic generation technique for measuring combinations of third-order elastic constants of cubic single crystals previously has been limited by the plane wave approximation to measurement of samples whose cross sectional dimensions are  $\geq 1.5$  cm. By obtaining data with small transducers and considering the effects of diffraction, the range of samples measurable with the technique has been extended to samples of  $\sim 5$  mm on a side. This development has made possible measurement of single crystals unavailable in larger dimensions. The minimum length required for measurement depends on the material and on the cross sectional dimensions; the current estimate of the minimum length is  $\sim 4$  mm. Diffraction corrections based on the Kirchhoff diffraction theory are presented and analyzed for both the fundamental frequency component and the second harmonic component of the ultrasonic wave. The technique was used to measure combinations of third-order elastic constants of small samples of  $\text{CsCdF}_3$  and  $\text{KZnF}_3$ .

The author expresses his sincere appreciation to Professor M. A. Breazeale for his suggestion of the topic and guidance during the research. He is grateful to Dr. W. T. Yost for his help in familiarizing him with the experimental procedure. Thanks are extended to Professor A. Zarembovitch of the Université Pierre et Marie Curie, Paris, France, for loaning the  $\text{CsCdF}_3$  and  $\text{KZnF}_3$  samples, and to Mr. D. W. Fitting for

providing the computer program for [D<sub>L</sub>]. He would like to thank Mrs. Maxine Martin for typing the manuscript.

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He is especially grateful to his parents for their love and encouragement.

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## CHAPTER I

### INTRODUCTION

The general objective of the research described here is to measure combinations of third-order elastic constants of cubic single crystals. In the macroscopic theory of elasticity, the strain energy density of the crystal can be expressed as a power series expansion in terms of the elements of the strain tensor. The coefficients of the second-order terms, multiplied by  $(2!)$ , are called second-order elastic constants; the third-order elastic constants are the coefficients of the third-order terms, multiplied by  $(3!)$ .

In the microscopic theory, the potential energy of the crystal can be expressed as a Taylor series in terms of the displacements of the atoms from their equilibrium positions. The coefficients in this series are called coupling parameters.

Pfleiderer (1962) and Coldwell-Horsfall (1963) expressed the second- and third-order coupling parameters for face-centered cubic crystals in terms of the second- and third-order elastic constants, assuming nearest neighbor interactions only. Coldwell-Horsfall (1963) presented similar relations for body-centered cubic crystals, assuming nearest and next-nearest neighbor interactions, and examined the case of central forces for both lattice types. Measurements of third-order elastic constants can therefore give information about the interatomic potential and forces between atoms.

Several physical properties of solids result from, or depend upon, the presence of the anharmonic terms in the strain energy density. These properties include thermal expansion, heat conduction, the proportionality of specific heat to temperature at temperatures above the characteristic Debye temperature, the difference between isothermal and adiabatic elastic constants, the dependence of second-order elastic constants on temperature and pressure, the interaction of two lattice waves, the attenuation of high frequency sound waves, and waveform distortion of sound waves.

Several experimental techniques have been used to measure third-order elastic constants of various solids. Measurement of the dependence of the second-order elastic constants on hydrostatic pressure gives combinations of third-order elastic constants but does not give the complete set of these constants for a particular crystal structure. Lazarus (1949) measured the pressure dependence of the second-order elastic constants of single crystals of NaCl, KCl, CuZn, Cu, and Al. Daniels and Smith (1958) made similar measurements on Cu, Ag, and Au. Hearmon (1953), using equations obtained by Birch (1947), determined what combinations of third-order elastic constants could be calculated from Lazarus' data.

Seeger and Buck (1960) and Melngailis, Maradudin, and Seeger (1963) proposed an optical method of measuring third-order elastic constants of transparent crystals, and Parker, Kelly, and Bolef (1964) used the method to measure a third-order elastic constant ( $C_{111}$ ) of NaCl. An initially sinusoidal finite amplitude ultrasonic wave propagating through the crystal distorts as it propagates, producing an asymmetric diffraction pattern of monochromatic light directed perpendicular to the

direction of the sound wave. Combinations of third-order elastic constants can be calculated from measurements of the light intensities in the positive and negative diffraction orders.

Barker and Hollenbach (1970) developed an interesting technique for measuring third- and higher-order elastic constants. The sample is struck by a projectile (and destroyed), and the resulting strain is measured. Graham (1972) calculated elastic constants of fused silica and sapphire from the data of Barker and Hollenbach (1970) and Graham and Brooks (1971).

One of the most widely used methods of measuring third-order elastic constants requires a measurement of the change in sound velocity with uniaxial stress. The first determination of a complete set of third-order elastic constants of isotropic materials was made by Hughes and Kelly (1953) by measuring the change of ultrasonic velocity with hydrostatic pressure and with uniaxial stress in polystyrene, iron, and pyrex glass. For the case of cubic crystals, Seeger and Buck (1960) obtained relations for the sound velocities in crystals subjected to hydrostatic pressure and uniaxial stress, in terms of second- and third-order elastic constants. Bateman, Mason, and McSkimin (1961) then performed the measurements on the cubic crystal germanium and obtained the first complete set of six third-order elastic constants of a cubic crystal. Measurements followed by Drabble and Gluyas (1963) on germanium, McSkimin and Andreatch (1964) on germanium and silicon, Bogardus (1965) on germanium, magnesium oxide, and fused silica, Chang (1965) on NaCl and KCl, and Thurston, McSkimin, and Andreatch (1966) on quartz. The third-order elastic constants obtained

by this method are neither truly adiabatic nor isothermal because an adiabatic wave propagates through an isothermally stressed medium (Skove and Powell, 1967).

To apply the above technique to metals would require small stresses because metal single crystals are easily deformed plastically. Hiki and Granato (1966) used a sensitive method of measuring changes in velocity with stress to measure complete sets of six third-order elastic constants of prestressed single crystals of copper, silver, and gold.

Another approach to the determination of third-order elastic constants involves the nonlinear phenomenon of the interaction of elastic waves. Jones and Kobett (1963) performed classical calculations of the scattering of two plane, intersecting, elastic waves in an anisotropic medium. Their predicted scattered waves were experimentally detected by Rollins (1963). Taylor and Rollins (1964) presented a quantum mechanical treatment of the process, and their results were compared with experimental measurements by Rollins, Taylor, and Todd (1964). Dunham and Huntington (1970) extended the theory of Taylor and Rollins and performed an experimental study using samples of fused silica and NaCl.

In the present experiments, combinations of third-order elastic constants are measured by an ultrasonic harmonic generation technique. As an initially sinusoidal wave propagates through a solid, higher harmonics are generated. This effect was observed and first reported by Gedroits and Krasilnikov (1963) and Breazeale and Thompson (1963). Breazeale and Ford (1965) established a relationship between the

third-order elastic constants and the solution to the nonlinear equation describing the propagation of an initially sinusoidal ultrasonic wave along a pure longitudinal mode direction in a cubic crystal. By measuring the amplitudes of the fundamental and second harmonic components, certain combinations of third-order elastic constants can be calculated, as described in Chapter II. A capacitive receiver capable of measuring the absolute amplitudes of the fundamental and second harmonic components of a plane wave was developed by Gauster and Breazeale (1966), and they used the receiver to measure combinations of third-order elastic constants of copper. Yost and Breazeale (1973) then used the capacitive receiver for measurements on fused silica and combined their results with the results of Dunham and Huntington (1970) to obtain the first complete set of adiabatic third-order elastic constants.

The harmonic generation technique has been valuable for low temperature measurements. Meeks and Arnold (1970) used the technique to measure combinations of third-order elastic constants of strontium titanate between 106 and 300°K, after Mackey and Arnold (1969) had performed the measurements at room temperature. Peters, Breazeale, and Paré (1968) developed a pneumatically controlled variable gap capacitive receiver to be used for low temperature measurements, and they used it to measure combinations of third-order elastic constants of copper down to liquid nitrogen temperature (Peters, Breazeale, and Paré, 1970). Similar measurements were performed on germanium (Yost and Breazeale, 1974). Bains and Breazeale (1976) then extended the technique through liquid helium temperature and measured germanium, and temperature

dependent measurements followed on four types of fused silica (Cantrell and Breazeale, 1978), on copper (Yost, Cantrell, and Breazeale, 1980), and on silicon (Philip and Breazeale, 1981).

In previous experiments, the measurements were performed on single crystals that were large enough to allow the use of transducers whose diameter to wavelength ratio permitted a plane wave approximation. However, some single crystal samples are not available in large sizes. To perform the measurements on smaller samples would require smaller diameter transducers. With a smaller diameter to wavelength ratio the plane wave approximation might no longer be valid and diffraction effects would need to be considered. The objectives of the present research were to develop the capability to perform the measurements on small samples, to present a theoretical model consistent with the results, and to use the method to make the measurements on a small sample.

A theory treating the generation of the second harmonic component in the field of a plane circular piston radiating into a fluid has been presented by Ingenito and Williams (1971). They solved a nonlinear wave equation by means of first-order perturbation theory to obtain an expression for the second harmonic as a function of position, in terms of an integral of the square of the fundamental component. They presented an expression for the value of the second harmonic integrated over a circular area coaxial with the source and of the same diameter as the source, and an expression for the second harmonic on the axis. Rogers (1970) presented a theory on the same topic. Kunitsyn and Rudenko (1978) treated the problem by solving the nonlinear equation



describing diffraction in the quasioptical approximation using the method of successive approximations. Lockwood, Muir, and Blackstock (1973) approached the problem by using weak-shock theory and restricted their analysis to the far (Fraunhofer) field.

In the present experiments, in order to develop the capability to perform the measurements on small samples, data were initially taken on large samples of copper. Combinations of third-order elastic constants of copper had been previously measured in the same laboratory by the harmonic generation technique, allowing an opportunity for comparison of the results of the present experiments with previous results. The small samples on which measurements were made were two members of the perovskite family,  $\text{CsCdF}_3$  and  $\text{KZnF}_3$ . Professor A. Zarembowitch and others at the Université Pierre et Marie Curie, Paris, France, have been studying structural phase transitions between the cubic and tetragonal structure in fluoperovskites and attempting to establish a connection between these instabilities and the nonlinear behavior of the materials. The data obtained in the present experiments complement their data, which is reported by Fischer (1979) and Fischer, Zarembowitch, and Breazeale (1981).

## CHAPTER II

### THEORY OF ULTRASONIC NONLINEARITY IN SOLIDS

The theory of ultrasonic nonlinearity in solids can be treated using classical continuum mechanics or quantum mechanics. A treatment using continuum mechanics will be presented in this chapter. The quantum mechanical approach gives identical results [cf. the tutorial paper by Bajak and Breazeale (1980)]. In the classical treatment the equations of motion are obtained from Lagrange's equations for continuous media. The potential energy term in the Lagrangian density is the strain energy density which is written in terms of the Lagrangian strains. The resulting equations of motion are simplified and solved. The solution shows that combinations of third-order elastic constants can be obtained by measurement of displacement amplitudes.

#### I. DEFINITION OF LAGRANGIAN STRAINS

Let the position of a point in an unstrained solid be given by the coordinates  $(a_1, a_2, a_3)$ . When the solid is strained, let the position of the point be given by the coordinates  $(x_1, x_2, x_3)$ . The components of the displacement are

$$u_i = x_i - a_i . \quad (II.1)$$

The subscript  $i$  can have the value 1, 2, or 3.

The square of the distance between two points that are close together is

$$d\ell^2 = da_i^2, \quad (II.2)$$

using the Einstein summation convention of summing over repeated indices. The square of the distance between the two points in the strained state is

$$\begin{aligned} d\ell'^2 &= dx_i^2 = (da_i + du_i)^2 = (da_i + \frac{\partial u_i}{\partial a_j} da_j)^2 \\ &= da_i^2 + 2 \frac{\partial u_i}{\partial a_j} da_i da_j + \frac{\partial u_i}{\partial a_j} \frac{\partial u_i}{\partial a_k} da_j da_k. \end{aligned} \quad (II.3)$$

Since  $d\ell'^2 = da_i^2$ , the above equation is

$$\begin{aligned} d\ell'^2 - da_i^2 &= 2 \left[ \frac{1}{2} \left( \frac{\partial u_i}{\partial a_j} + \frac{\partial u_j}{\partial a_i} + \frac{\partial u_k}{\partial a_i} \frac{\partial u_k}{\partial a_j} \right) \right] da_i da_j \\ &= 2\eta_{ij} da_i da_j. \end{aligned} \quad (II.4)$$

The  $\eta_{ij}$  are the elements of the Lagrangian strain tensor.

## II. THE EQUATIONS OF MOTION

The equations of motion can be obtained from Lagrange's equations for continuous media (Holt and Ford, 1967). Lagrange's equations are

$$\frac{d}{dt} \left( \frac{\partial L}{\partial \dot{x}_i} \right) + \frac{d}{da_k} \left( \frac{\partial L}{\partial (x_i/a_k)} \right) = 0. \quad (II.5)$$

The Lagrangian density is

$$L = \frac{1}{2} \rho \dot{x}_i^2 - \phi(\eta) , \quad (\text{II.6})$$

where  $\rho$  is the unstrained mass density and  $\phi(\eta)$  is the strain energy per unit unstrained volume. The properties of the medium enter the equations of motion through the strain energy density  $\phi(\eta)$ .

### III. THE ELASTIC CONSTANTS

The strain energy density can be written as a power series expansion about the state of zero strain in terms of the Lagrangian strains:

$$\phi(\eta) = \phi_0 + \phi_1 + \phi_2 + \phi_3 + \dots . \quad (\text{II.7})$$

The first term,  $\phi_0$ , represents the energy of the unstrained medium and is constant; the second term,  $\phi_1$ , is zero because the first derivative of  $\phi$  with respect to strain, evaluated at zero strain, is equal to zero. The terms of order higher than three are neglected. The terms that will be of interest in Lagrange's equations can be written, using the Einstein summation convention,

$$\phi_2 = \frac{1}{2!} C_{ijkl} \eta_{ij} \eta_{kl} , \quad (\text{II.8})$$

and

$$\phi_3 = \frac{1}{3!} C_{ijklmn} \eta_{ij} \eta_{kl} \eta_{mn} . \quad (\text{II.9})$$

The coefficients  $C_{ijkl}$  appearing in the second-order terms are called

the second-order elastic constants and the  $C_{ijklmn}$  are called the third-order elastic constants.

The elastic constants can also be defined with a thermodynamic approach if the physical process being considered is purely adiabatic or purely isothermal. Denoting the internal energy per unit volume by  $U$  and the Helmholtz free energy per unit volume by  $F$ , the elastic constants are defined (Brugger, 1964) by

$$C_{ijkl\dots}^S = \left\{ \frac{\partial^n U}{\partial \eta_{ij} \partial \eta_{kl} \dots} \right\}_S \quad (\text{adiabatic, } n \geq 2) \quad (\text{II.10})$$

and

$$C_{ijkl\dots}^T = \left\{ \frac{\partial^n F}{\partial \eta_{ij} \partial \eta_{kl} \dots} \right\}_T \quad (\text{isothermal, } n \geq 2), \quad (\text{II.11})$$

where  $S$  denotes the entropy and  $T$  denotes the temperature.

The elastic constants defined by Eq. (II.10) are called adiabatic elastic constants, and the elastic constants defined by Eq. (II.11) are called isothermal elastic constants. Since  $F = U - TS$ , the distinction does not apply at absolute zero.

The propagation of an ultrasonic wave through an unstrained medium is an adiabatic process. To describe this process, the strain energy density  $\phi$  appearing in the Lagrangian density is identified with the internal energy density  $U$ . Thus, the quantities measured in the present experiments are combinations of adiabatic third-order elastic constants.

In general there are 81 second-order and 729 third-order elastic constants. The number of independent constants is lower if there are lattice symmetries. For all cubic crystals there are three independent second-order elastic constants, and for the most symmetrical classes of cubic crystals there are six independent third-order elastic constants (Birch, 1947). All of the samples measured in the present experiments were cubic crystals having six independent third-order elastic constants.

It is conventional and convenient to contract the subscript notation for the elastic constants (Voigt, 1928) as follows:

$$11 \rightarrow 1, 22 \rightarrow 2, 33 \rightarrow 3, 23 \rightarrow 4, 13 \rightarrow 5, 12 \rightarrow 6.$$

Since the strain tensor elements  $\epsilon_{ij}$  are symmetric, permutations of the subscripts  $ij$  are equivalent. The subscript notation for the strains will not be contracted. Using Brugger's (1964) notation, the three independent second-order elastic constants for cubic crystals are  $C_{11}$ ,  $C_{12}$ , and  $C_{44}$ , and the six independent third-order elastic constants are  $C_{111}$ ,  $C_{112}$ ,  $C_{123}$ ,  $C_{144}$ ,  $C_{166}$ , and  $C_{456}$ .

#### IV. THE NONLINEAR WAVE EQUATION AND ITS SOLUTION

The nonlinear wave equation describing the propagation of an ultrasonic wave through the sample can be obtained from Lagrange's equations (II.5). Identifying  $\phi(\eta)$ , appearing in the Lagrangian density (II.6), with the internal energy per unit volume  $U$ , as previously discussed, the following equation can be obtained:

$$\rho \ddot{x}_i = \frac{d}{da_k} \left[ \frac{\partial U}{\partial n_{ik}} + \frac{\partial U}{\partial n_{qk}} \frac{\partial u_i}{\partial a_q} \right]. \quad (\text{II.12})$$

This equation applies for any direction of propagation and any wave polarization. It can be simplified by considering only pure mode longitudinal plane waves. Pure longitudinal waves can propagate in the [100], [110], and [111] directions in a cubic crystal. The equation of motion pertaining to a pure longitudinal plane wave propagating in one of these three directions becomes (Breazeale and Ford, 1965)

$$\rho \ddot{u} = K_2 \frac{\partial^2 u}{\partial a^2} + (3K_2 + K_3) \frac{\partial u}{\partial a} \frac{\partial^2 u}{\partial a^2}, \quad (\text{II.13})$$

where  $K_2$  and  $K_3$  are linearly independent combinations of second-order and third-order elastic constants, respectively, as shown in Table II-1.

The perturbation solution (valid for distances much less than the discontinuity distance) to Eq. (II.13) is given by Breazeale and Ford (1965) for a sinusoidal driver located at  $a = 0$  by

$$u = A_1 \sin(ka - \omega t) - \left( \frac{3K_2 + K_3}{8K_2} \right) A_1^2 k^2 a \cos 2(ka - \omega t) + \dots, \quad (\text{II.14})$$

where

$A_1$  is the displacement amplitude of the fundamental frequency component at  $a = 0$ ,

$k$  is the wavenumber,

$a$  is the distance of propagation,

$\omega$  is the angular frequency,

$t$  is time.

Table II-1.  $K_2$  and  $K_3$  for the Principal Directions in a Cubic Crystal

Direction	$K_2$	$K_3$
[100]	$C_{11}$	$C_{111}$
[110]	$\frac{C_{11} + C_{12} + 2C_{44}}{2}$	$\frac{C_{111} + 3C_{112} + 12C_{166}}{4}$
[111]	$\frac{C_{11} + 2C_{12} + 4C_{44}}{3}$	$\frac{1}{9} (C_{111} + 6C_{112} + 12C_{144} + 24C_{166} + 2C_{123} + 16C_{456})$



The first term in the solution is the component having the fundamental frequency; the second term is the component having the second harmonic frequency. The solution also contains higher harmonic terms which have been neglected. The amplitude of the second harmonic component, denoted by  $A_2$ , is given by

$$A_2 = -\left(\frac{3K_2 + K_3}{8K_2}\right)A_1^2 k^2 a, \quad (\text{II.15})$$

and is seen to be proportional to the square of the fundamental amplitude, the square of the frequency, the propagation distance, and a term containing the combinations of elastic constants. By measuring the displacement amplitudes of the fundamental and second harmonic components and knowing the value of  $K_2$ , the combinations of third-order elastic constants  $K_3$  can be calculated. The value of  $K_2$  for a given direction is given by

$$K_2 = \rho v^2 \quad (\text{II.16})$$

where  $v$  is the velocity of sound in the given direction. Rearranging Eq. (II.15) gives

$$K_3 = -K_2\left(3 + \frac{8A_2}{A_1^2 k^2 a}\right) = -K_2(3 + R). \quad (\text{II.17})$$

The second term in the parentheses in Eq. (II.17) is denoted by  $R$ :

$$\beta = \frac{8A_2}{A_1^2 k_a^2} = -\left(\frac{3K_2 + K_3}{K_2}\right) . \quad (\text{II.18})$$

The quantity  $\beta$  is equal to the negative of the ratio of the coefficient in the nonlinear term in the wave equation (II.13) to the coefficient in the linear term;  $\beta$  is therefore called the nonlinearity parameter. (This definition of the nonlinearity parameter  $\beta$  is consistent with the corresponding definition for liquids and gases. The expression given for  $\beta$  is a factor of three greater than the expression used for  $\beta$  by Cantrell (1976).)

The theory presented in this chapter has led to an equation that shows that the nonlinearity parameter  $\beta$  (and  $K_3$ ) can be determined from a measurement of the amplitudes of the first and second harmonic components of a plane wave. Two of the objectives of the present research are to develop the capability to measure these quantities using waves that are not plane, and to form a better understanding of the diffraction of an ultrasonic wave propagating in a nonlinear medium. A comparison of the values to be measured with accepted values of  $\beta$  and  $K_3$  serves as a basis for formulating an approach to nonlinear diffraction theory.

## CHAPTER III

### APPARATUS, PROCEDURE, AND SAMPLES

#### I. EXPERIMENTAL CONSIDERATIONS

The method of measuring the displacement amplitudes of the fundamental and second harmonic components of a plane ultrasonic wave propagating through a solid sample will be discussed in this chapter. For the experiments to be described, as well as for previous experiments [Gauster (1966) and others], a fundamental frequency of 30 MHz was selected for the following reasons. The amplitude of the second harmonic component is proportional to the square of the fundamental frequency; therefore the higher the frequency, the better the signal to noise ratio. However, attenuation and the effects of nonparallelism of the surfaces of the sample also increase with increasing frequency. A frequency of 30 MHz was found to be a good compromise. Also, the diameters of the transducers used in previous measurements were large enough (1.27 cm) that the diameter to wavelength ratio using 30 MHz was large enough to justify a plane wave approximation.

The ultrasonic wave was pulsed in order to avoid interference effects. A pulse repetition rate of 60 pulses/second was used to minimize heating of the sample.

In the theory of finite amplitude wave propagation the second harmonic amplitude is related to the fundamental amplitude by a simple integral power law only in the limit of infinitesimal fundamental

amplitude (Thurston and Shapiro, 1967). Therefore the initial fundamental amplitudes were minimized to levels providing useful signal to noise ratios for the generated second harmonic components.

## II. THE DETECTOR APPARATUS

The measurements of the displacement amplitudes were made using the detector apparatus pictured in Figure III-1. A cross sectional view of the apparatus is shown in Figure III-2. An ultrasonic pulse is transmitted into the sample from a piezoelectric x-cut quartz transducer and causes the optically flat end of the sample to vibrate. An optically flat electrode is located approximately ten microns below the end of the sample and forms parallel plate capacitor with it. A dc bias voltage of the order of 150 volts is applied across this capacitor. When the ultrasonic pulse impinges upon the end of the sample, the gap spacing of the capacitor changes, and a current is produced. The fundamental frequency signal and the second harmonic signal are separated electronically. The displacement amplitudes  $A_1$  and  $A_2$  are calculated by determining the current and the initial gap spacing.

The initial gap spacing is measured with an impedance bridge. The current is determined by measuring the voltage across a known impedance. Since it is preferable to measure a continuous voltage rather than a pulsed signal, a substitutional continuous signal is used which is adjusted to produce the same output signal as that received using the pulsed ultrasonic wave.

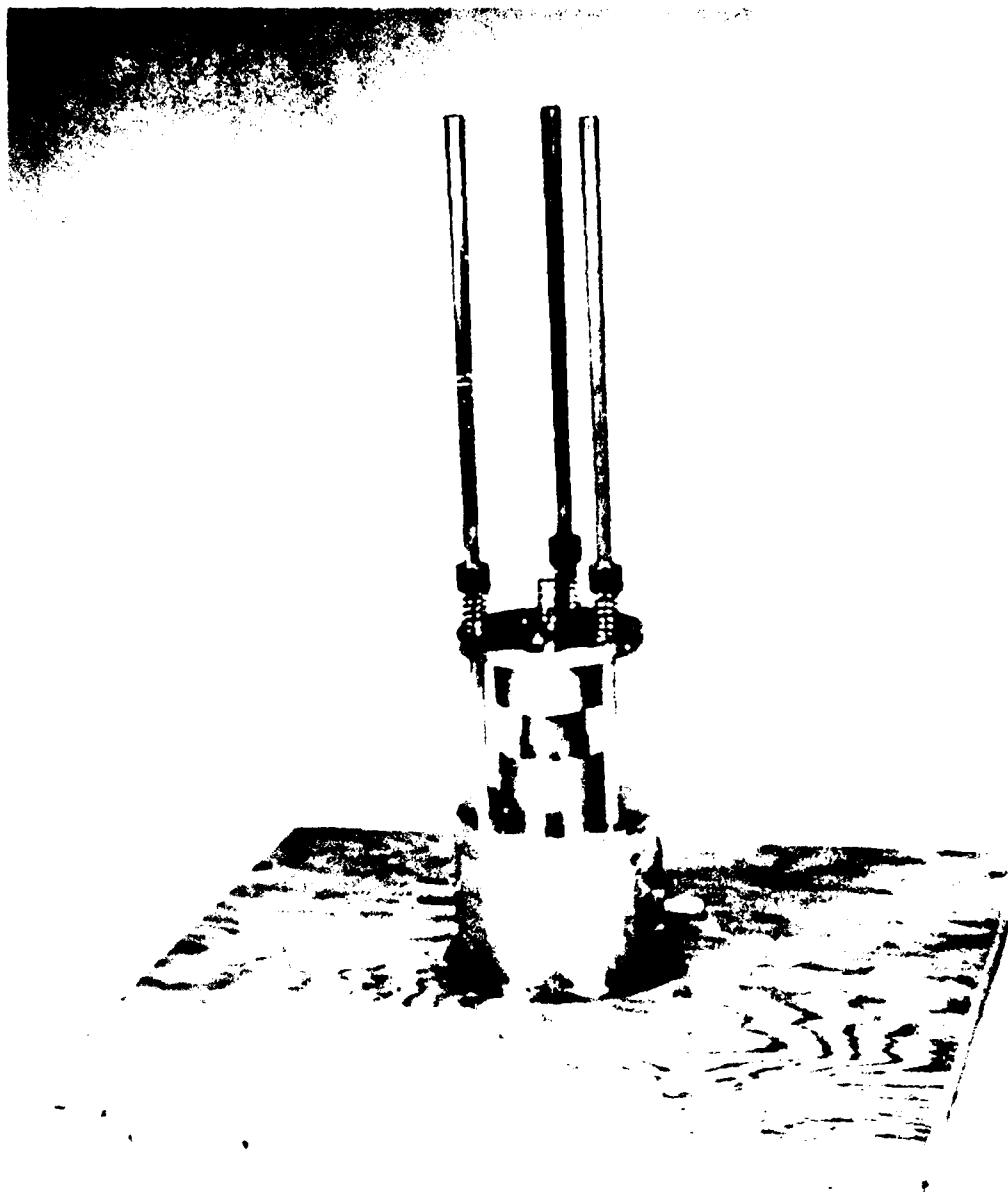


Figure III-1. The detector apparatus. A 2.5 cm diameter sample is shown.

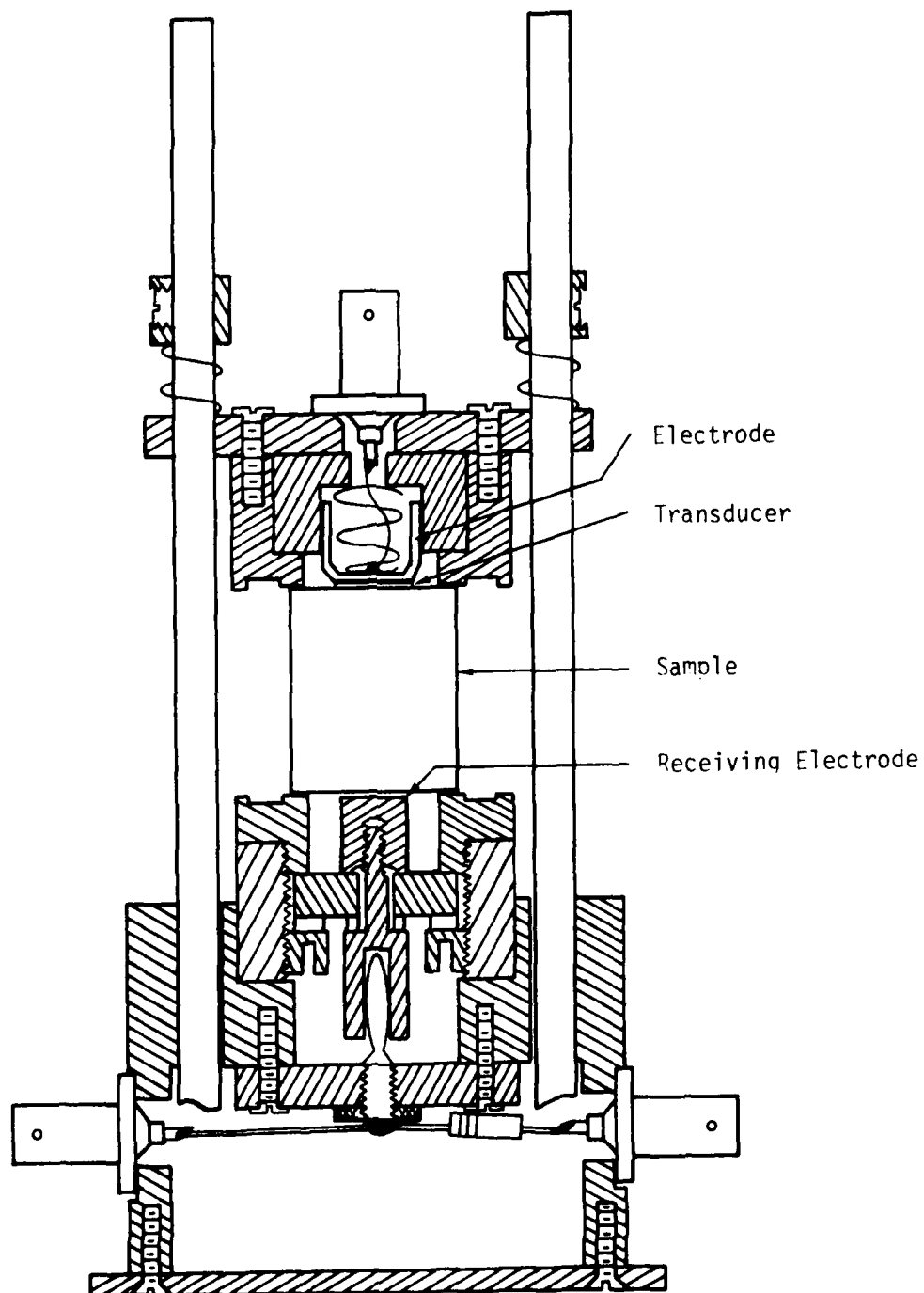


Figure III-2. Cross sectional view of the detector apparatus.

### III. CALIBRATION AND PROCEDURE

The circuit diagram used for calculating the amplitudes is shown in Figure III-3. The detector circuit is represented by a Norton equivalent circuit. The circuit elements and parameters are defined as follows.

$C_D$  is the quiescent capacitance of the detector,

$C_S$  is the stray capacitance of the detector,

$L$  is the inductance of the wire leading from the banana jack to the BNC connector (shown in Figure III-2).

$Z$  is the impedance of the resistor located in the base of the apparatus, indicated in Figure III-2,

$G_D$  is the current generator of the Norton equivalent circuit of the detector,

$G_S$  is the substitutional signal current generator,

$i_D$  is the amplitude of the current produced by the ultrasonic wave,

$i_S$  is the amplitude of the substitutional current,

$S_1$  is a "switch" that is opened and closed by turning on or off the ultrasonic pulse,

$S_2$  is a "switch" that is opened and closed by disconnecting or connecting a signal generator.

Peters (1968) showed that the Thévenin equivalent circuit of the capacitive receiver is a voltage source of voltage amplitude

$$V = \frac{2AV_b}{S_0}, \quad (\text{III.1})$$

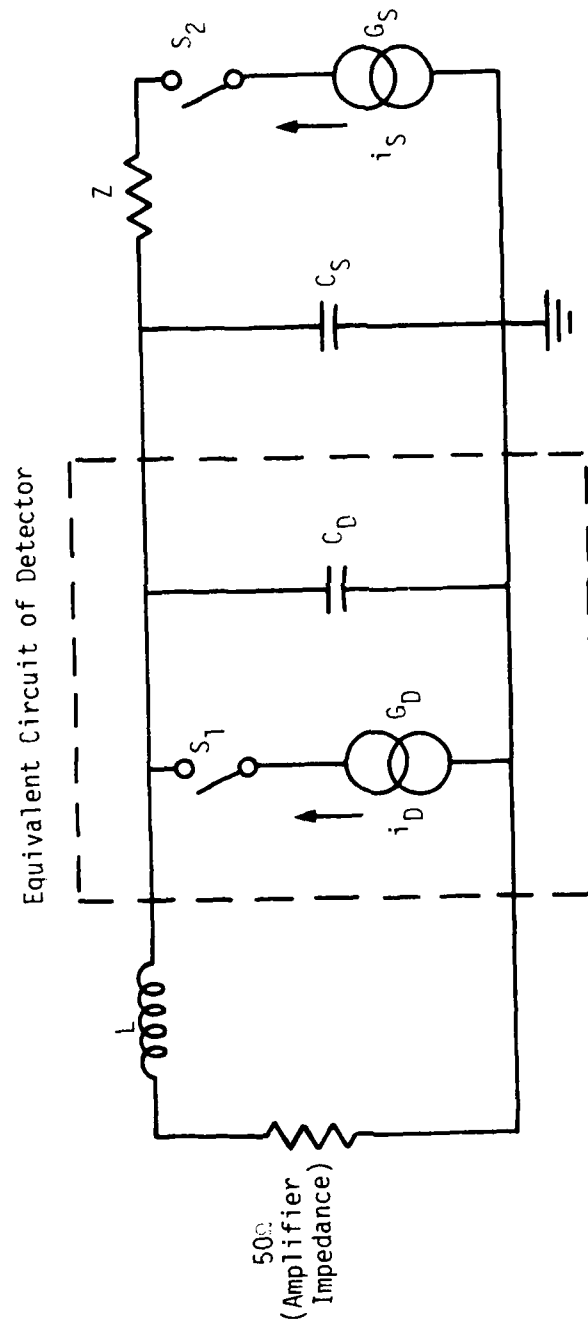


Figure III-3. Equivalent circuit used for the displacement amplitude measurements.



in series with a capacitor  $C_D$ , feeding the amplifier impedance. In Eq. (III.1),  $A$  is the displacement amplitude of the ultrasonic wave within the solid,  $V_b$  is the dc bias voltage applied across the capacitive receiver, and  $S_0$  is the gap spacing of the quiescent capacitive receiver. The amplitude of vibration of the free surface of the sample is twice the amplitude within the sample because the incident and reflected waves add. This was taken into account in obtaining Eq. (III.1).

The current amplitude  $i_D$  produced by the capacitive detector is related to the voltage amplitude  $V$  by

$$i_D = V\omega C_D, \quad (\text{III.2})$$

where  $\omega$  is the angular frequency of vibration. Combining Eqs. (III.1) and (III.2) gives

$$i_D = \frac{2AV_b\omega C_D}{S_0}. \quad (\text{III.3})$$

The output from the substitutional source  $G_S$  is adjusted to give the same output from the amplifier as the output resulting from the ultrasonic pulse. Under this condition,  $i_D$  is equal to  $i_S$ , and the displacement amplitude  $A$  can be calculated from Eq. (III.3) by measuring  $i_S$ .

The current  $i_S$  is determined by measuring the voltage  $V_S$  across the current generator  $G_S$  and measuring the impedance through which  $i_S$  passes. The equation is

$$i_S = \frac{V_S}{|Z + [j\omega(C_D + C_S) + \frac{1}{50 + j\omega L}]^{-1}|} \quad (III.4)$$

The quantity  $Z$  is the impedance of the resistor located between the substitutional source and the capacitive detector. At the frequencies used in these experiments, the impedance of the resistor is not purely resistive and is a function of frequency. The impedance can be determined by the following procedure. Refer to Figure III-4 which shows the circuit used for the measurement of the impedance  $Z$ . The sample, detector assembly, and bottom plate are removed from the apparatus, and  $50 \, \Omega$  terminators are connected to the two BNC connectors at the base of the apparatus. A cw variable-frequency signal generator is connected to the terminator on the side having the resistor. Probes A and B of a (Hewlett-Packard 8405A) vector voltmeter are positioned at point 1, the phase angle between the signals is zeroed, and the voltages are measured. Probe B is then moved to point 2, leaving probe A at point 1. The signal generator is adjusted to give the same channel A voltage as before, and the channel B voltage and the phase angle are measured. The impedance  $Z$  is calculated from the formula

$$Z = [j\omega C + \frac{1}{R_1 + j\omega L}]^{-1} \frac{V_{B1} - V_{B2}e^{j\phi}}{V_{B2}e^{j\phi}}, \quad (III.5)$$

where  $C$  is the stray capacitance at point 2, including the probe tip capacitance,  $R_1$  is the resistance of the precision terminator measured with an impedance bridge,  $V_{B1}$  and  $V_{B2}$  are the voltages measured by

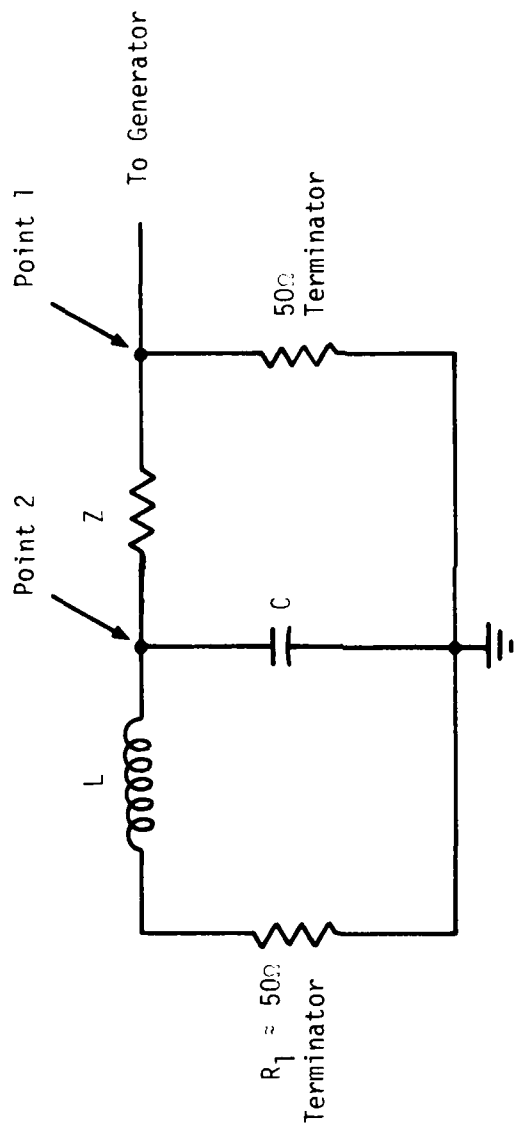


Figure III-4. Circuit used for the measurement of the impedance  $Z$ .

probe B at points 1 and 2, respectively, and  $\phi$  is the phase angle. The other symbols were defined previously.

The quantities  $C$ ,  $R_1$ , and  $L$  were measured. The value of  $L$  was  $3.8 \times 10^{-8}$  H; the value of  $R_1$  for the particular terminator used was  $49.74 \Omega$ . The value of the stray capacitance  $C$  depends on the configuration during the voltage measurements; in the present measurement  $C$  was measured to be  $10.75 \times 10^{-12}$  F. Measurements of the impedance  $Z$  were performed at the five frequencies from 28.0 MHz to 32.0 MHz in increments of 1.0 MHz and at the five frequencies from 56.0 MHz to 64.0 MHz in increments of 2.0 MHz. A computer program was written which used the Lagrange five point interpolation formula (Abramowitz and Stegun, 1964) to calculate  $|Z|$  as a function of frequency from 28.00 MHz to 32.00 MHz in increments of 0.01 MHz and from 56.00 MHz to 64.00 MHz in increments of 0.02 MHz.

The magnitude of  $Z$  was about 11 k $\Omega$  at the fundamental frequency and about 9 k $\Omega$  at the second harmonic frequency. The quantity in brackets in Eq. (III.4) can be neglected compared to  $Z$  at the frequencies used in these experiments. Therefore Eq. (III.4) can be approximated as

$$i_S = \frac{V_S}{|Z|} . \quad (\text{III.6})$$

Equating  $i_D$  given by Eq. (III.3) with  $i_S$  given by Eq. (III.6) gives an equation for the displacement amplitude:

$$A = \frac{V_S S_0}{2V_b \omega C_D |Z|} . \quad (\text{III.7})$$

The quantity  $A_2/A_1^2$ , which appears in the nonlinearity parameter  $B$ , can be written

$$\frac{A_2}{A_1^2} = \left( \frac{V_{S2}}{V_{S1}} \right)^2 \frac{V_{b\omega_1} C_D |Z_1|^2}{S_0 |Z_2|}, \quad (\text{III.8})$$

where the subscripts 1 and 2 refer to the fundamental and second harmonic, respectively.

A block diagram of the experimental arrangement used for the measurements of the displacement amplitudes is shown in Figure III-5. A cw signal from a variable-frequency oscillator (VFO) is separated into pulses by a gated amplifier; the pulses pass through a 30 MHz bandpass filter and drive the piezoelectric quartz transducer. The pulsed signal from the capacitive receiver is sent through either a 30 MHz or 60 MHz bandpass amplifier which isolates the signal to be measured, rectifies the signal, and yields an output that is the envelope of the rectified pulse. The VFO and gated amplifier are tuned to optimize the shape and amplitude of this envelope. The output signal from the amplifier is monitored with an oscilloscope and measured with a boxcar integrator, the output of which is proportional to the time average of the input. Therefore random noise is averaged to zero while the repetitive signal adds to produce a measurable output voltage. Then the substitutional signal discussed previously is applied to the capacitive receiver and adjusted by means of attenuators and an amplifier gain control to produce the same output as that obtained from the ultrasonic pulse, as measured by the boxcar integrator. To obtain the substitutional signal at the second

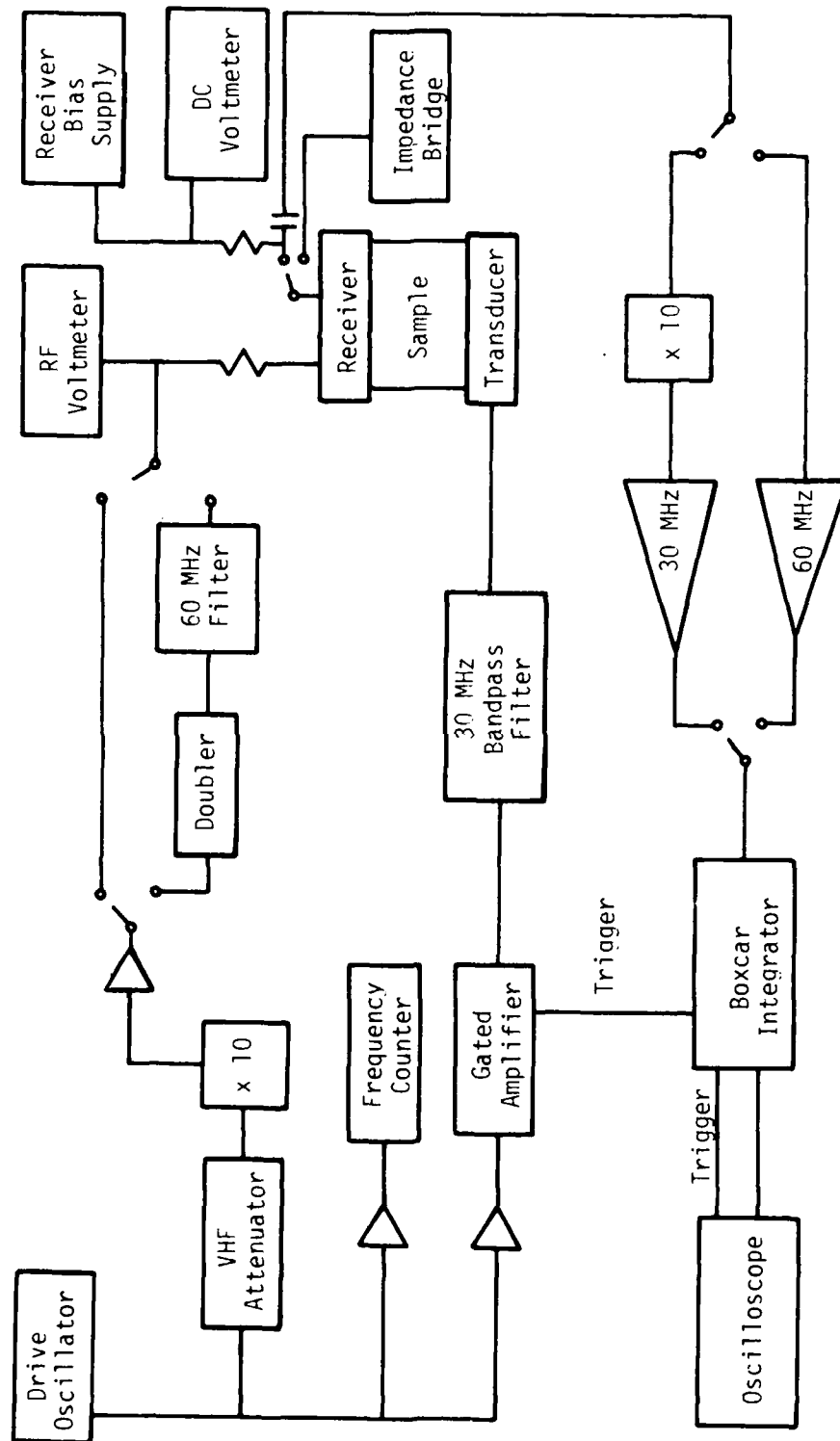


Figure III-5. Block diagram of the experimental arrangement used for the displacement amplitude measurements.

harmonic frequency, the signal at the fundamental frequency is doubled by a ring bridge mixer and filtered with a 60 MHz bandpass filter. The (RMS) voltage across the signal generator is measured with an rf voltmeter to determine  $V_S$ .

#### IV. THE QUARTZ TRANSDUCERS AND RECEIVING ELECTRODES

To investigate the effects of smaller diameter transducers and receiving electrodes on the amplitude measurements, several quartz transducers and receiving electrodes of various diameters were made. The diameters used in the experiments are listed on p. 35. For each of the ten electrodes of diameters 1.27-0.188 cm, a corresponding ground ring was made having inside diameter 0.1 inch (2.54 mm) greater than the diameter of the electrode. The brass receiving electrodes and ground rings were machined using standard techniques and hand lapped. The quartz transducers were reduced to the desired diameter by grinding the edge of a larger transducer (or a broken fragment if available) with a high speed ( $\sim 20,000$  rpm) silicon carbide grinding wheel. See Figure III-6. The transducer is secured to the end of a rod by attaching to the rod the adhesive from a cellulose tape in a manner similar to that described by Yost (1972, p. 36). (The polyethylene film is not necessary.) The quartz crystal is then very carefully pressed onto the end of the rod. Then the rod, whose end surface was machined to be perpendicular to the rod's axis, is placed in a collet in a milling machine. The grinding wheel is secured to the table of the milling machine. The rod and quartz crystal are slowly rotated by hand as the wheel grinds the quartz. After each revolution of the quartz crystal,

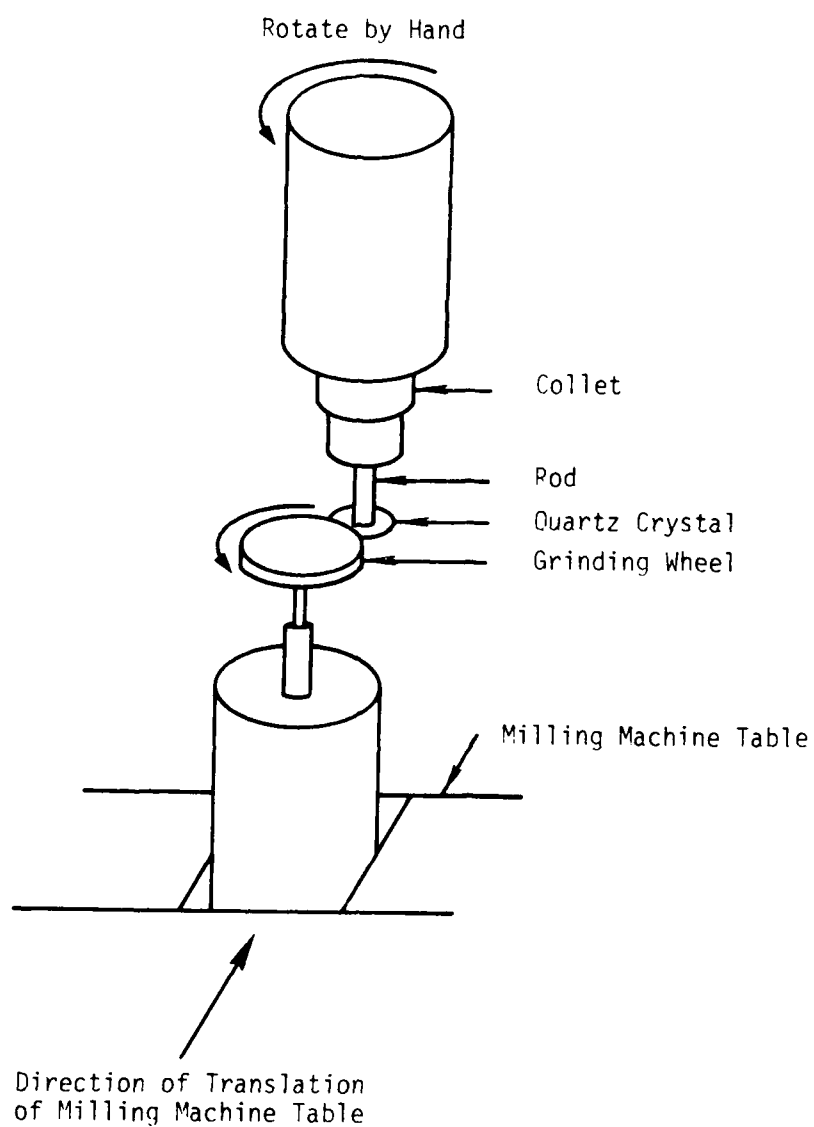


Figure III-6. Diagram illustrating the procedure for grinding the quartz crystal transducers.

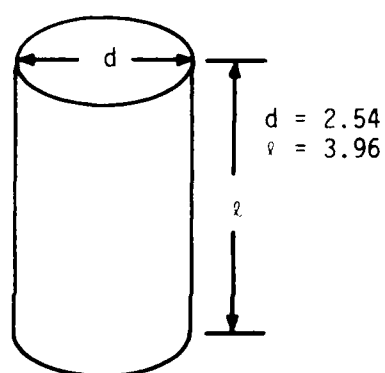


the grinding wheel is moved slightly into the quartz using the translational motion of the table. This procedure is repeated until the desired diameter is obtained. Care should be taken in removing the quartz crystal from the rod. It may be necessary to dissolve the adhesive using a solvent such as benzene.

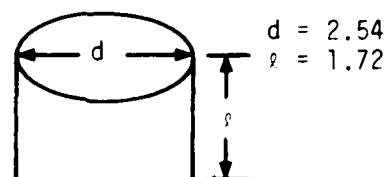
## V. THE SAMPLES

The samples on which measurements were made included [100] and [111] copper samples, a sample of potassium zinc fluoride ( $\text{KZnF}_3$ ) and a sample of cesium cadmium fluoride ( $\text{CsCdF}_3$ ), each of which had faces perpendicular to the [100] and [110] directions, and a [111]  $\text{CsCdF}_3$  sample. (The [111] Cu sample was used in nonlinearity measurements performed by Gauster (1966) and Peters (1968). Gauster and Peters also reported measurements on a [100] Cu sample different from the one used in the present experiments.) All of the samples are cubic crystals; copper has the face-centered cubic structure, and  $\text{CsCdF}_3$  and  $\text{KZnF}_3$  have the perovskite structure. The copper samples had been neutron irradiated to pin the dislocations, which reduces nonlinear effects of the dislocations. The dimensions, shapes, and crystallographic directions are shown in Figure III-7. The densities and the values of  $K_2$  are given in Table III-1. The  $K_2$  values for copper were calculated using the second-order elastic constants of Overton and Gaffney (1955):

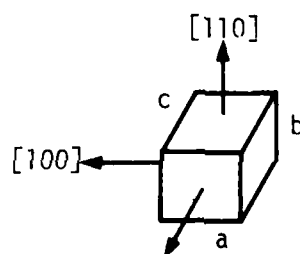
$$\begin{array}{lll} C_{11} = 1.684 & C_{12} = 1.214 & C_{44} = 0.754 \\ \text{(in units of } 10^{12} \text{ dynes/cm}^2\text{).} & & \end{array}$$



A. Cu [111]

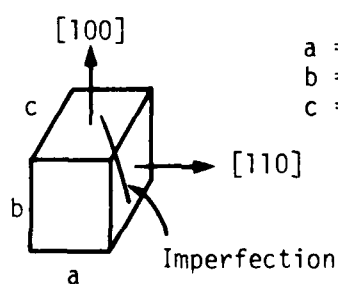


B. Cu [100]



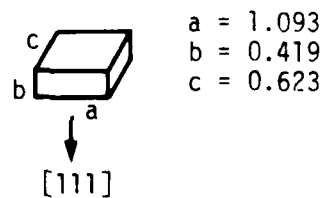
a = 1.203  
b = 1.039  
c = 1.017

C.  $\text{KZnF}_3$



a = 1.170  
b = 1.388  
c = 1.197

D.  $\text{CsCdF}_3$  [100] and [110]



a = 1.093  
b = 0.419  
c = 0.623

E.  $\text{CsCdF}_3$  [111]

Figure III-7. Drawings of the samples. The dimensions are given in centimeters.

Table III-1. Sample Densities and  $K_2$  Values

Sample	Density ( $10^3$ kg/m <sup>3</sup> )	Direction	$K_2$ ( $10^{12}$ dynes/cm <sup>2</sup> )
Cu	8.95	[100]	1.684
		[111]	2.376
CsCdF <sub>3</sub>	5.638	[100]	1.08
		[110]	0.995
		[111]	0.967
KZnF <sub>3</sub>	4.02	[100]	1.345
		[110]	1.317

The surfaces at the transducer and receiver ends of each sample were lapped by hand optically flat. The  $\text{CsCdF}_3$  and  $\text{KZnF}_3$  samples should not be touched with bare hands; they were handled using laboratory gloves. It is permissible, however, to lap these materials with an oil slurry lapping compound. This should be done carefully because the surfaces are easily scratched.

To provide a conducting surface at the transducer and receiver ends of the  $\text{CsCdF}_3$  and  $\text{KZnF}_3$  samples, a layer of copper of the order of 1000 Å thick was evaporated onto the surfaces.

The quartz transducers were bonded to all samples with Nonaq Stopcock Grease.

An imperfection of approximately ten millimeters in length exists in the ([100] and [110])  $\text{CsCdF}_3$  sample as indicated in Figure III-7.

## VI. THE SEQUENCE OF THE EXPERIMENTAL INVESTIGATION

The first step taken to investigate the effects of smaller diameter transducers and receivers was to obtain data on the Cu [111] sample using four different diameter transducers with a 0.636 cm diameter receiving electrode. The diameters used for these data and the other data mentioned in this section are listed in Table III-2. Data were then obtained on the same sample using a 1.18 cm diameter receiving electrode and three different diameter transducers. Next, data were taken on the Cu [100] sample using the 0.636 cm diameter receiving electrode and four different diameter transducers. Data were then taken using transducers and receiving electrodes of equal diameter. The measurements were made on the Cu [100] sample using ten diameters.

Table III-2. Transducer and Receiver Diameters Used in the Experiments

Sample	Direction	Length (cm)	Receiver Diameter (cm)	Transducer Diameter (cm)
Cu	[111]	3.96	0.636	1.27
				0.540
				0.370
				0.195
	[100]	1.72	1.18	1.27
				0.540
				0.370
				0.189
			0.636	1.27
				0.562
				0.370
				0.189
				1.27
				1.14
				1.00
				0.889
				0.764
				0.635
				0.508
				0.381
				0.254
				0.188
CsCdF <sub>3</sub>	[100]		0.508	
			0.381	
	[110]		0.508	
			0.381	
	[111]		0.381	
KZnF <sub>3</sub>	[100]	0.508		
	[110]	0.381		

After determining a method for correcting the data for diffraction, measurements on  $\text{CsCdF}_3$  and  $\text{KZnF}_3$  were performed.

## CHAPTER IV

### RESULTS, DISCUSSION, AND THEORETICAL TREATMENT OF DIFFRACTION

To perform the third-order elastic constant measurements on small samples requires small diameter transducers and receivers. It was anticipated that the waves emitted from the smaller transducers would not satisfy the plane wave assumption, and the results of the measurements would differ from results obtained with large diameter transducers. In order to determine how the smaller transducers and receivers affect the measurements, and to learn how to make a reliable determination of third-order elastic constants from measurements made with small diameters, data were taken as outlined in Chapter III, Section VI.

#### I. RESULTS OF THE MEASUREMENTS WITH UNEQUAL DIAMETER TRANSDUCERS AND RECEIVING ELECTRODES

The results of the initial measurements on the Cu [111] sample using two different receiving electrode sizes and a range of transducer sizes are plotted in Figures IV-1 and IV-2. The quantities  $V_1$  and  $V_2$  are the voltmeter readings of the fundamental and second harmonic voltages, respectively, and are equal to the root-mean-square values of the input voltages. If a plane wave were incident over the full face of the receiving electrode, according to Eq. (III.8) the quantity  $V_2/V_1^2$  would be proportional to  $A_2/A_1^2$  for a given bias voltage, frequency, and receiver diameter and gap spacing. Eq. (II.18) shows that

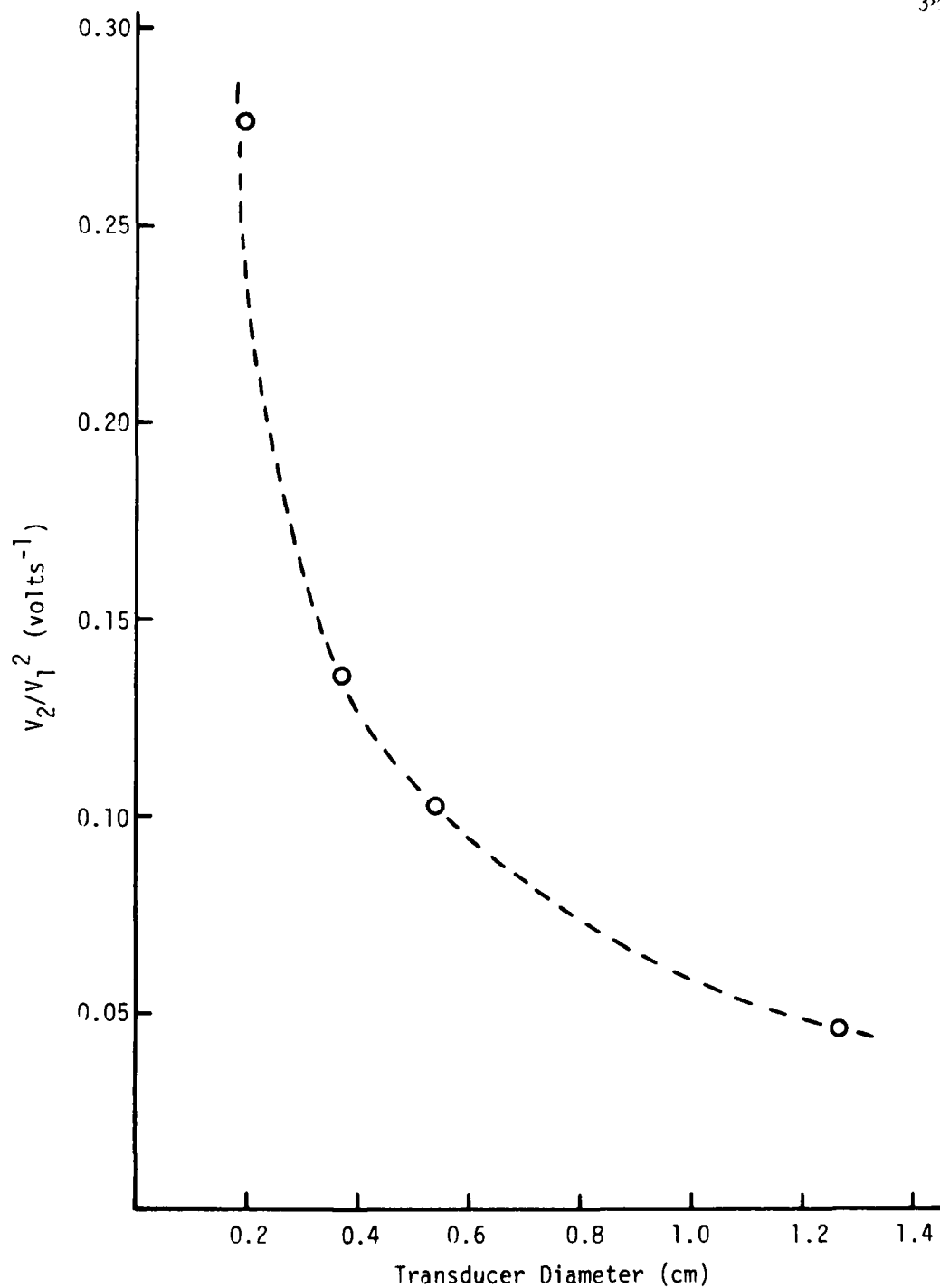


Figure IV-1.  $V_2/V_1^2$  versus transducer diameter for the 3.95 cm Cu [111] sample using the 0.636 cm diameter receiving electrode.



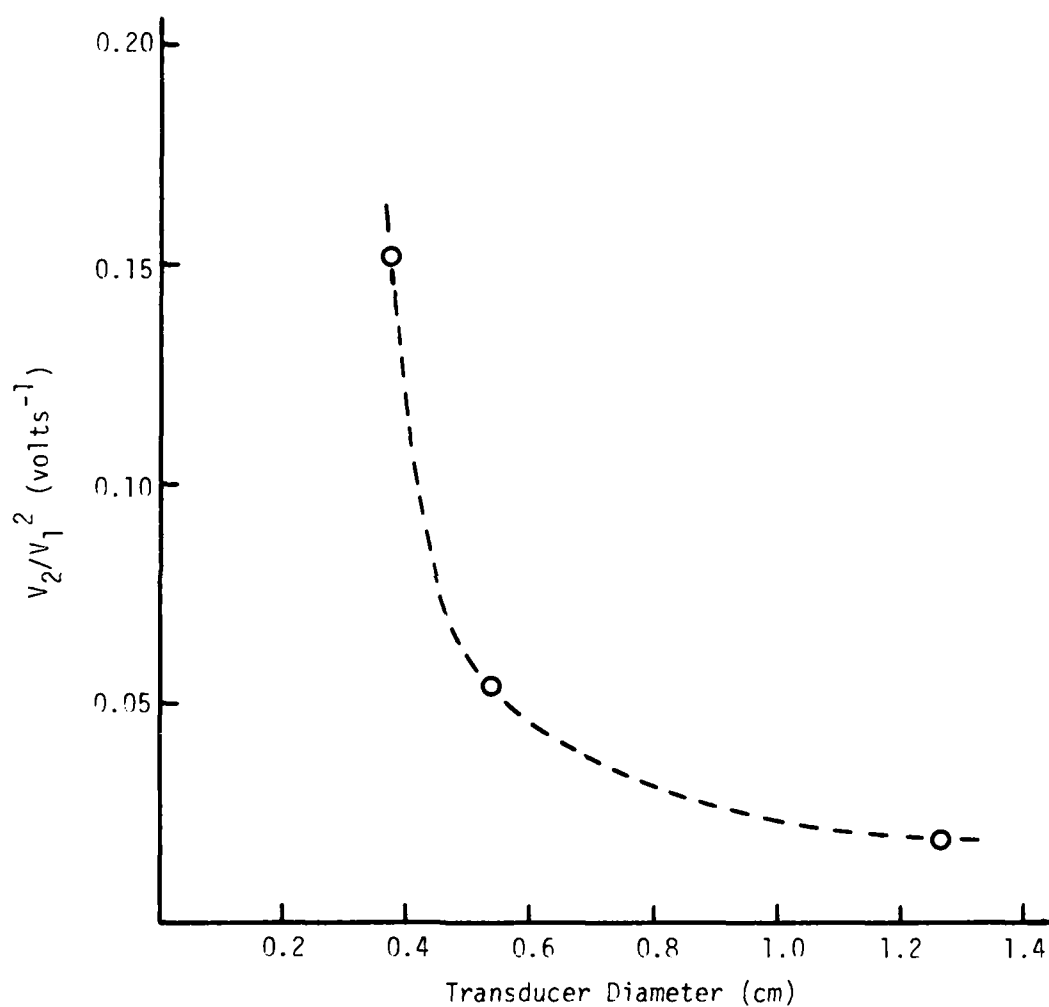


Figure IV-2.  $V_2/V_1^2$  versus transducer diameter for the 3.96 cm Cu [111] sample using the 1.18 cm diameter receiving electrode.

the quantity  $A_2/A_1^2$  should be constant for a given substance for a given frequency, sample length, and direction of propagation of the ultrasonic wave. Figures IV-1 and IV-2 show that the measured values of  $V_2/V_1^2$  depend strongly on transducer diameter for a given receiver diameter. Slight differences in bias voltage, frequency, and gap spacing for the data points on a given curve would not significantly alter the curves. The results of the next set of measurements, on the Cu [100] sample of length 1.721 cm, are shown in Figure IV-3. The curve, obtained using a shorter sample and different direction of wave propagation, is flatter than the two previously discussed curves but still indicates a difference between the measurements with the smallest transducer and with the larger transducers.

Measurements performed with 12.7 mm diameter transducers have been shown to yield accepted values of the nonlinearity parameter  $\beta$ . The  $V_2/V_1^2$  values in Figures IV-1 through IV-3 tend to decrease smoothly with transducer diameter and approach these accepted results. Therefore, curves such as those in Figures IV-1 through IV-3 can be used as calibration curves for obtaining the nonlinearity parameters for small samples. A family of curves for different sample lengths would be useful. To obtain the nonlinearity parameter for a given sample measured with a given diameter transducer, a correction factor would be obtained from the selected calibration curve by comparing the value on the curve at the given diameter with the value approached at the large-diameter end of the curve.

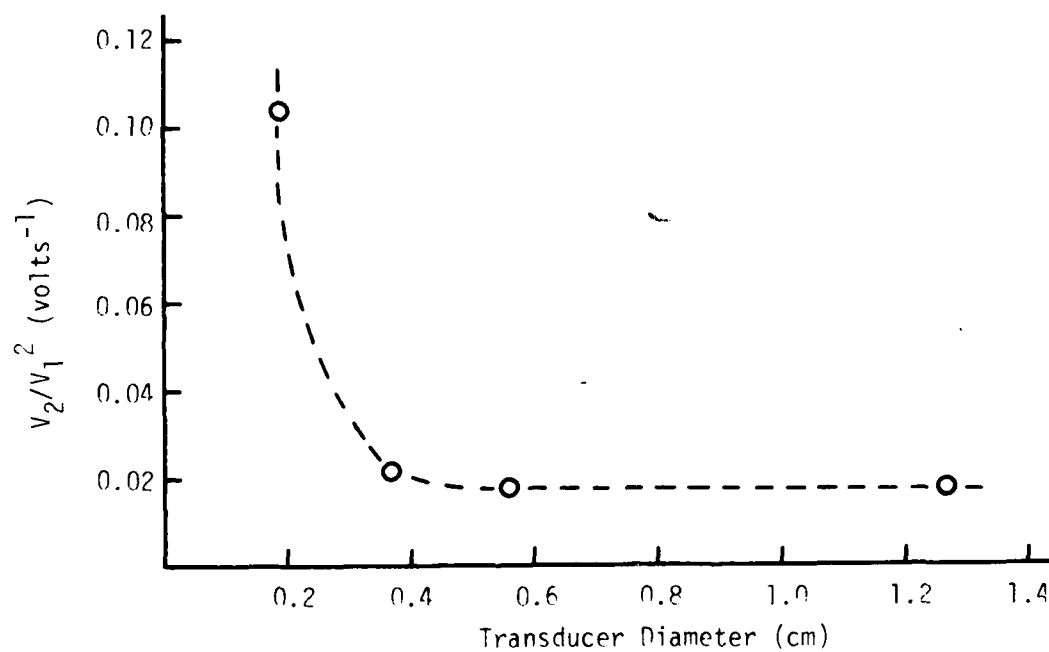


Figure IV-3.  $V_2/V_1^2$  versus transducer diameter for the 1.72 cm Cu [100] sample using the 0.636 cm diameter receiving electrode.

## II. RESULTS OF THE MEASUREMENTS WITH EQUAL DIAMETER TRANSDUCERS AND RECEIVING ELECTRODES

One of the objectives of the present research is to develop a theoretical model for the nonlinear diffraction problem that is consistent with the results. The results reported in the previous section are not optimum for a mathematical understanding of the problem. It was decided to obtain data using transducers and receiving electrodes of equal diameter. This would possibly facilitate the mathematical treatment of the problem and might allow the utilization of existing diffraction theory for that configuration.

Ten sets of data, using the diameters listed in Table III-2, p. 35, were taken on the 1.72 cm Cu [100] sample. For each of the ten sets of data, a measurement was taken for ten different values of the amplitude of the ultrasonic wave, with the exception of the measurements made with the 1.0 cm diameter transducer and receiver, in which the amplitude was varied through five values. Values of the fundamental amplitude and the second harmonic amplitude were calculated for each amplitude setting by assuming the wavefronts to have been plane, with no correction for diffraction. These values will be denoted by  $A_{1u}$  and  $A_{2u}$ , the subscript u meaning "uncorrected for diffraction." For each of the ten data sets a graph of  $A_{2u}$  versus  $A_{1u}^2 k^2 a$  was plotted. The graphs for the largest and smallest diameters used, 1.27 cm and 0.189 cm, are shown in Figures IV-4 and IV-5, respectively. The graphs for the other eight data sets are similar. The straight line represents a linear least squares fit to the data. Each graph verifies

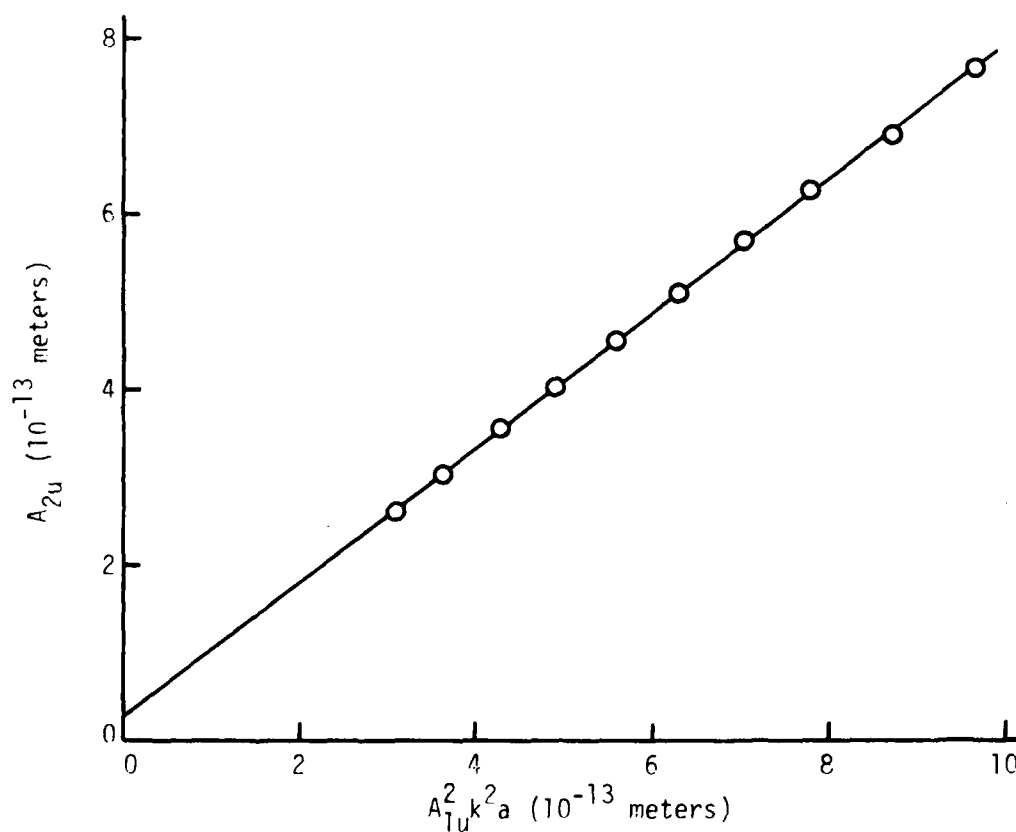


Figure IV-4.  $A_{2u}$  versus  $A_{1u}^2 k^2 a$  for Cu [100] using the 1.27 cm diameter transducer and receiving electrode.

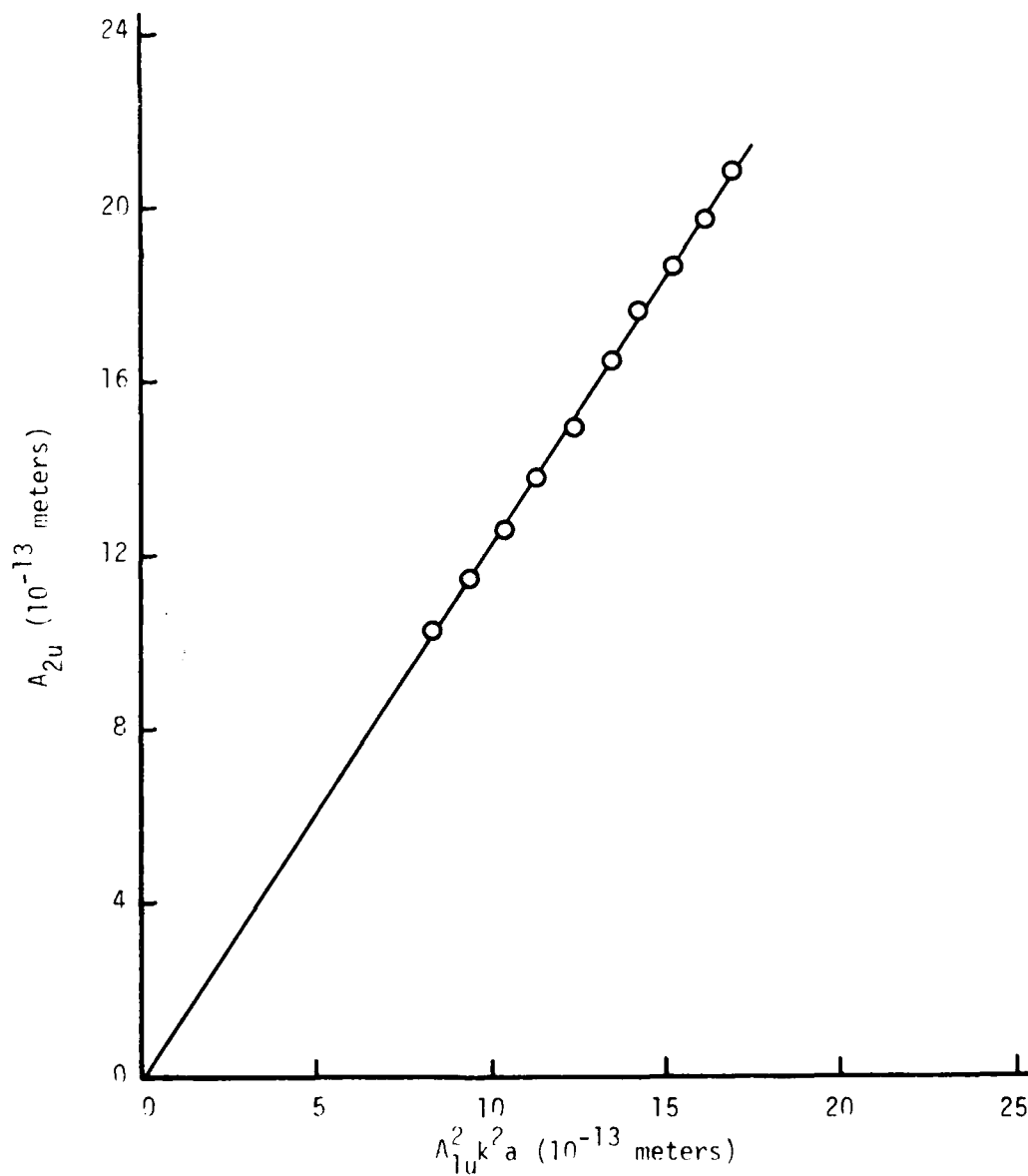


Figure IV-5.  $A_{2u}$  versus  $A_{1u}^2 k^2 a$  for Cu [100] using the 0.189 cm diameter transducer and receiving electrode.

the expected proportionality of the amplitudes of the second harmonic component to the square of the amplitude of the fundamental component. The straight lines do not pass through the origin. Bains (1974) has attributed this type of result to residual noise. The present values of  $A_{2u}$  were corrected by subtracting the value of the  $A_{2u}$  intercept, calculated from a least squares linear regression formula.

The values of the nonlinearity parameter ( $= 8A_{2u}/A_{1u}^2 k^2 a$ ) could have been calculated from the slopes of the lines in these graphs. However, Yost (1972) pointed out that a different method has the advantages of allowing measurements with good signal-to-noise ratios and yielding a value which satisfies the assumption of infinitesimal amplitude waves. In this method, the values of  $A_2/A_1^2$  (or  $\beta$ ) calculated for each data point are plotted as a function of  $A_1$ . A curve drawn through these data points is extrapolated to  $A_1 = 0$ , and the value at this point is used for the nonlinearity parameter  $\beta$ . This method has been particularly advantageous to previous experimenters because their data at higher values of  $A_1$  differed significantly from the data at lower values. However, the graphs of the calculated values of  $\beta_u$  versus  $A_{1u}$  for the present experiments, examples of which for the 1.27 cm and 0.189 cm diameter transducers and receivers are given in Figures IV-6 and IV-7, respectively, do not show a trend in the data as a function of  $A_{1u}$ . Therefore, for each data set the nonlinearity parameter  $\beta_u$  was taken to be the average of the individual values.

These values for the nonlinearity parameter, which were calculated with no correction for diffraction, are plotted as a function of

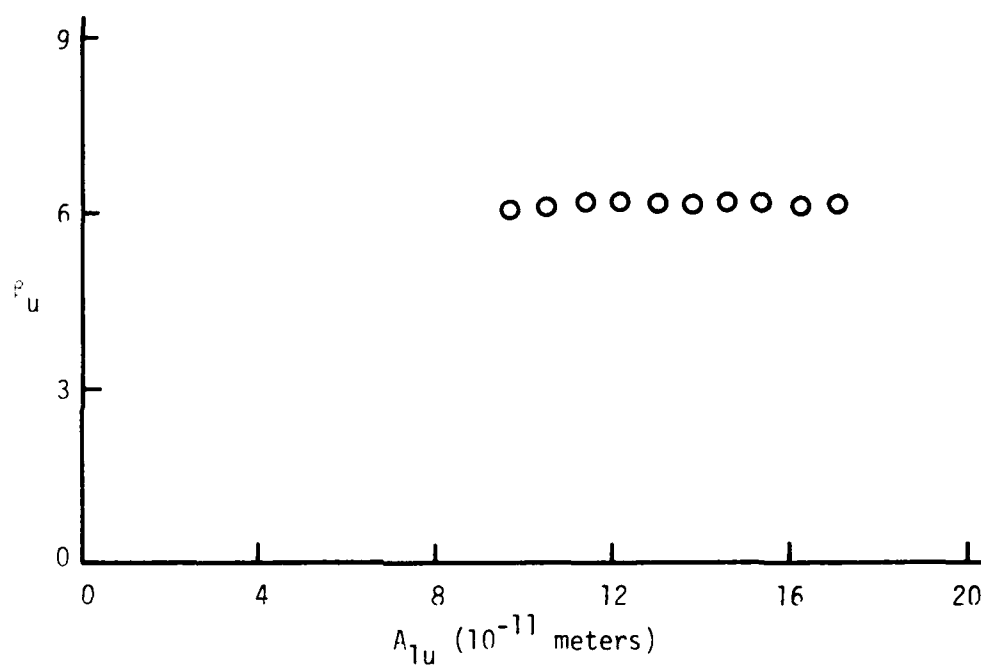


Figure IV-6.  $P_u$  versus  $A_{1u}$  for Cu [100] using the 1.27 cm diameter transducer and receiving electrode.



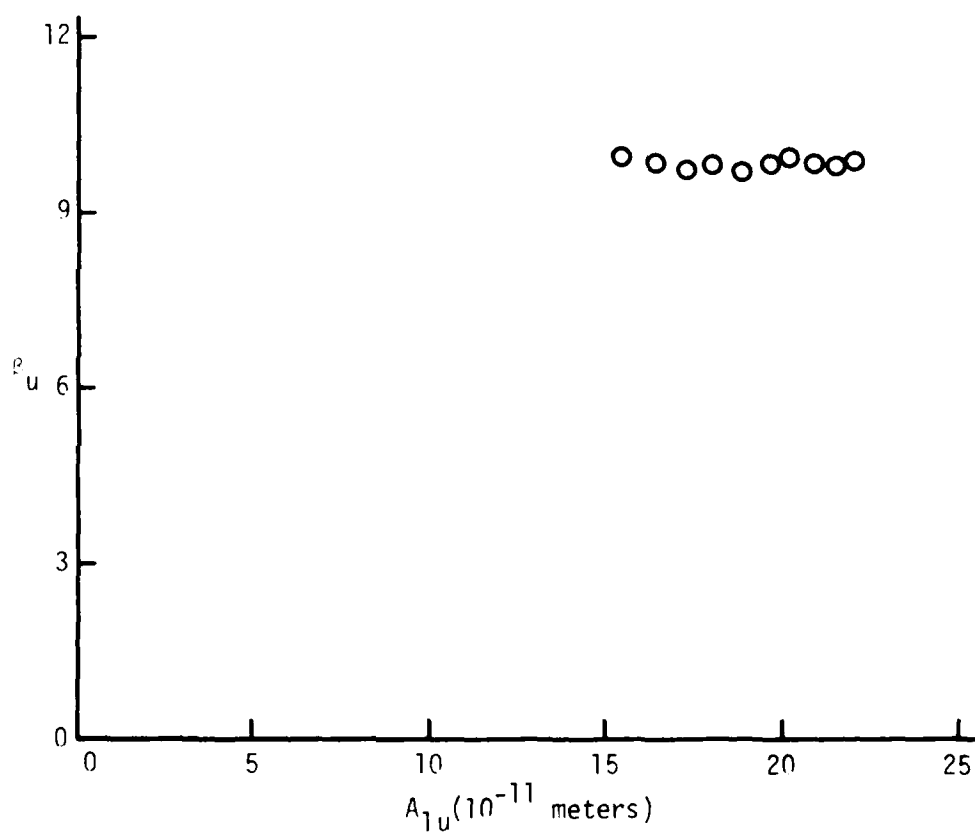


Figure IV-7.  $R_u$  versus  $A_{1u}$  for Cu [100] using the 0.189 cm diameter transducer and receiving electrode.

transducer and receiver diameter in Figure IV-8 and are listed on p. 53. (There was an experimental problem in obtaining the data with the 0.76 cm diameter transducer and receiver, and these data will not be included in the analysis.) These values of the nonlinearity parameter which were obtained using equal diameter transducers and receivers are more consistent as a function of transducer diameter than were the values obtained by varying the transducer diameter while keeping the receiver diameter fixed. However, these  $\beta_u$  values still are not constant as a function of diameter.

### III. THE DIFFRACTION CORRECTION

The effects of diffraction will now be considered. In Chapter II a theory was presented that enables one to calculate  $\beta$  and  $K_3$  from amplitude measurements of a plane wave field described by nonlinear acoustics. It is desired to be able to determine  $\beta$  and  $K_3$  for small samples requiring small diameter transducers that emit waves that are not plane, and to obtain a better understanding of the diffraction of this nonlinear wave field. In the experiments, a circular transducer oscillating at a particular frequency emits a wave into the medium. As the wave propagates, higher harmonic components are generated. (In the present study only the fundamental and second harmonic components are used in the calculations.) For a plane wave, the second harmonic component depends on the amplitude of the fundamental component, the frequency, and the distance of propagation as stated in Chapter II. For a wave that is not plane, it is presumed that the second harmonic

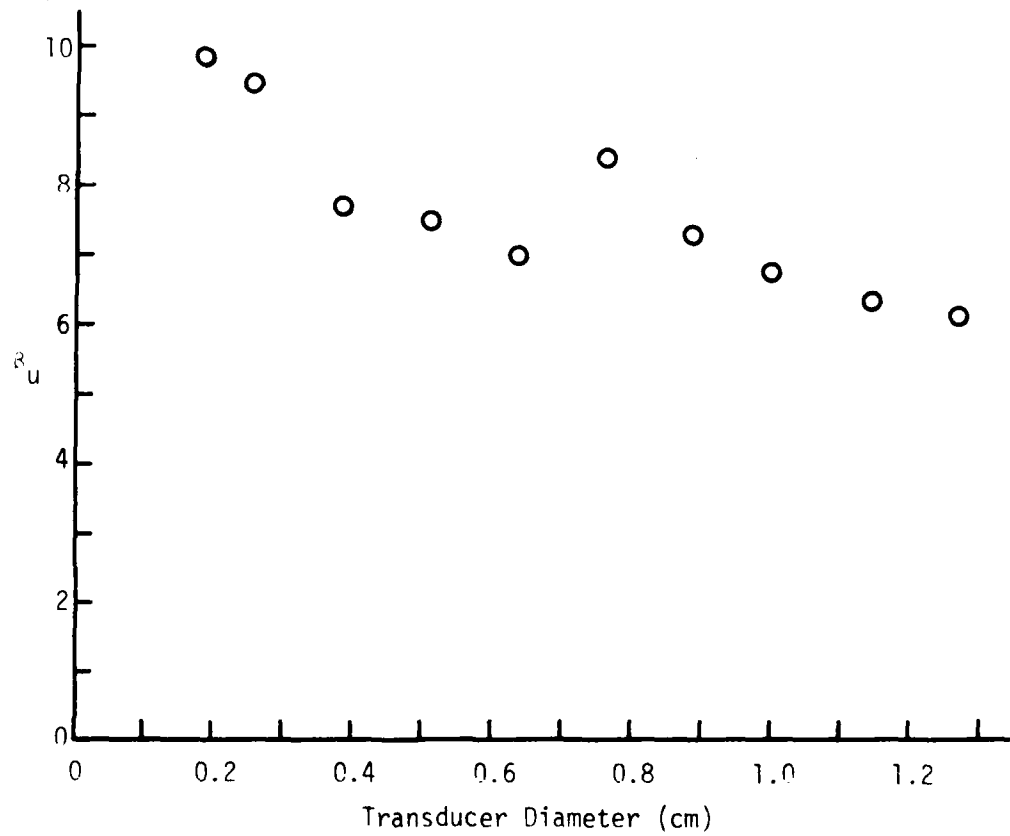


Figure IV-8.  $R_u$  versus transducer diameter for Cu [100].

also depends on these factors, but it is not known a priori what the dependence is or how to describe the diffracted nonlinear field. The diffraction correction that allows a determination of  $\beta$  and  $K_3$  is the subject of this section.

The diffraction of the fundamental component will be considered first, and the second harmonic component will be treated later. By neglecting the loss of energy of the fundamental component to the second harmonic, the diffraction of the fundamental component can be treated by linear acoustics theory. The theory to be applied to the fundamental component was obtained for a homogeneous fluid medium. Analysis of data obtained with an anisotropic solid medium by utilizing a diffraction theory applicable to a fluid medium is not unprecedented; cf. Seki, Granato, and Truell (1956). [Papadakis (cf. Mason and Thurston, 1975) extended the approach used by Seki et al. to application to anisotropic solids for the linear diffraction problem.]

It is assumed that the field produced by the circular transducer in the present experiments is the same as the field produced by a plane circular piston source, set in an infinite rigid baffle and oscillating at the fundamental frequency. The diffraction correction integral is defined as the integral of the field from the piston source over the surface of a circular area of diameter equal to that of the source and coaxial with the source, divided by the integral over the same area of a plane wave field considered to be produced by an infinitely extended plane source oscillating with the same amplitude and frequency as the piston. An approximate diffraction correction integral can be obtained by integrating an approximate expression for

the piston field given by Lommel (1886). This integral, called the Lommel diffraction correction integral, denoted by  $D_L$ , has been evaluated numerically or graphically by several authors. Rogers and Van Buren (1974) evaluated the integral analytically and obtained a simple closed-form expression for it, valid at all distances from the source provided that  $(kR)^{1/2} \gg 1$ . Their result for the magnitude of  $D_L$  is

$$|D_L| = \{[\cos(2\pi/s) - J_0(2\pi/s)]^2 + [\sin(2\pi/s) - J_1(2\pi/s)]^2\}^{1/2} \quad (\text{IV.1})$$

where

$J_0$  = the zero order Bessel function,

$J_1$  = the first order Bessel function,

$s = 2\pi z/kR^2 = z/(R^2/\lambda)$ ,

$z$  = the distance from the source plane to the field plane,

$k$  = the wavenumber,

$R$  = radius of the piston,

$\lambda$  = wavelength.

This expression for  $|D_L|$  was used for correcting the data for the diffraction of the fundamental component. For each data set the value of  $s$  was calculated and  $|D_L|_1$  was computed, the subscript 1 referring to the fundamental frequency component. Each nonlinearity parameter  $B_u$  was corrected by multiplying it by the square of  $|D_L|_1$  since  $A_1^2$  appears in the denominator of the expression for  $B$ . The corrected values are given by

$$\beta_{c,1} = |D_L|_1^2 \beta_u. \quad (\text{IV.2})$$

The resulting values are denoted by  $\beta_{c,1}$ , the subscript c meaning "corrected" and the subscript 1 meaning that it was the fundamental component that was corrected for diffraction. The values of the quantities used in the calculations and the corrected values of the nonlinearity parameters are given in Table IV-1, in which the subscript 1 indicates that the value refers to the fundamental component. The corrected values of the nonlinearity parameters are plotted as a function of transducer diameter in Figure IV-9.

By examining Figure IV-9 it is seen that for the first time in this study the results are consistent as a function of transducer diameter. A measure of the consistency is the percent deviation of the corrected nonlinearity parameters from the mean, which is tabulated in Table IV-1. The mean value,  $\bar{\beta}_{c,1}$ , is 5.536.

This mean value of the corrected nonlinearity parameter values can be used to calculate  $K_3$  by inserting  $\bar{\beta}_{c,1}$  into Eq. (II.17). The result is  $K_3 = (-14.4 \pm 0.3) \times 10^{12}$  dynes/cm<sup>2</sup>. Gauster and Breazeale (1968), using a transducer having a diameter large enough that a diffraction correction was not considered necessary, obtained the value  $K_3 = (-14.3 \pm 0.44) \times 10^{12}$  dynes/cm<sup>2</sup>, and Peters (1968) obtained  $K_3 = (-13.9 \pm 0.2) \times 10^{12}$  dynes/cm<sup>2</sup>. [Gauster and Breazeale performed the calculations using the second-order elastic constants given by Hiki and Granato (1966); Peters' results and the present results were calculated using the constants given by Overton and Gaffney (1955).] There

Table IV-1. Values of the Nonlinearity Parameter Corrected for Diffraction of the Fundamental Component and Quantities Used in the Calculation\*

Transducer Diameter (cm)	$S_1$	$ D_L _1$	$ D_L _1^2$	$\beta_u$	$\beta_{c,1}$	Percent Deviation From the Mean
1.27	0.0610	0.946	0.894	$6.134 \pm 0.039$	$5.484 \pm 0.035$	1
1.15	0.0747	0.941	0.885	$6.336 \pm 0.057$	$5.607 \pm 0.050$	1
1.01	0.0954	0.933	0.871	$6.733 \pm 0.171$	$5.864 \pm 0.149$	6
0.891	0.1236	0.924	0.854	$6.269 \pm 0.081$	$5.354 \pm 0.069$	3
0.640	0.2342	0.897	0.804	$6.986 \pm 0.075$	$5.617 \pm 0.060$	1
0.512	0.3736	0.870	0.756	$7.469 \pm 0.102$	$5.646 \pm 0.077$	2
0.385	0.6481	0.836	0.699	$7.694 \pm 0.054$	$5.378 \pm 0.038$	3
0.258	1.4434	0.748	0.559	$9.461 \pm 0.140$	$5.289 \pm 0.078$	4
0.189	2.6871	0.753	0.567	$0.851 \pm 0.081$	$5.585 \pm 0.046$	1

\*Each uncertainty given is one standard deviation using (N-1) weighting.

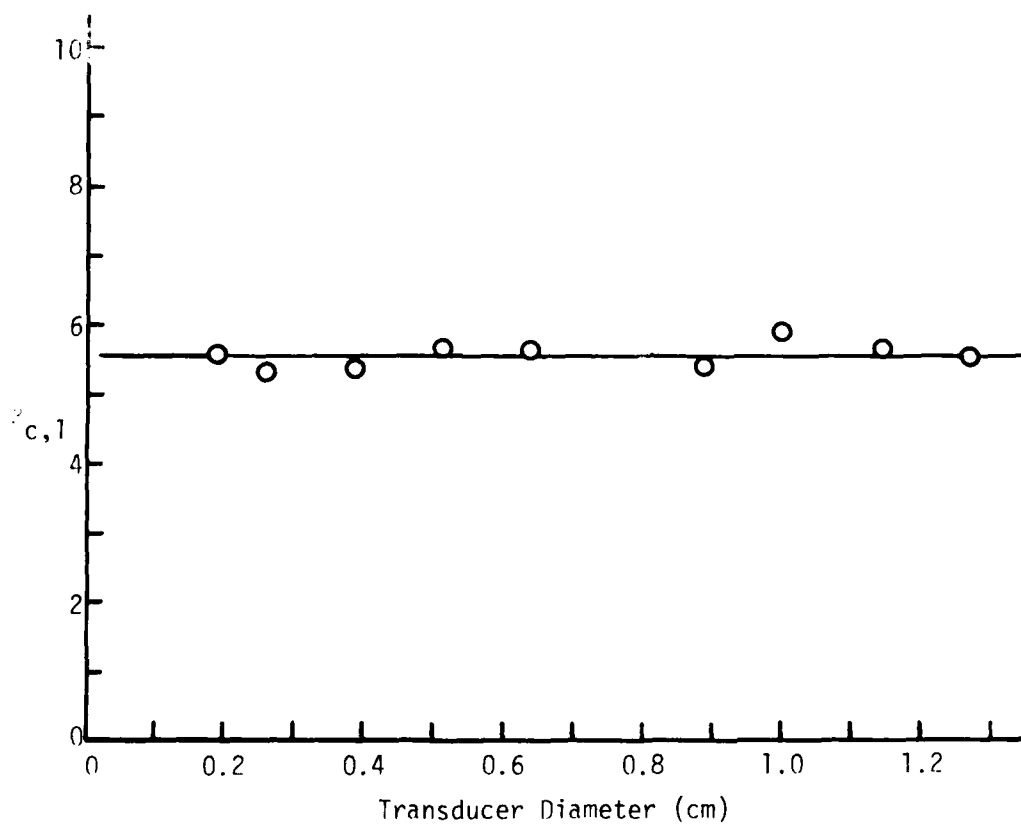


Figure IV-9.  $P_{c,1}$  versus transducer diameter for Cu [100]. The straight line represents the mean value  $\bar{P}_{c,1} = 5.536$ .



is very good agreement between the results of the present measurements and the previous results.

By using transducers and receivers of equal diameter and correcting the data for diffraction of the fundamental field by using the Lommel diffraction correction integral, results were obtained that were consistent as a function of diameter and agreed with previous measurements.

This conclusion is encouraging; however, no correction for diffraction of the second harmonic component has been applied. A model for the diffraction of the second harmonic field allowing a diffraction correction to be calculated will now be proposed. Essentially, the Kirchhoff diffraction theory has been applied to the fundamental component. Therefore, the initial wave field can be considered to be composed of spherical Huyghens wavelets of fundamental frequency emitted from elements over the surface of the source. Each spherical wavelet generates a second harmonic component. By symmetry, the generated second harmonic wavelets must also be spherical. The amplitude of the second harmonic wavelets depends on some function of distance. An integrated value over the receiver of this second harmonic field is measured, and from this measurement the second harmonic plane wave amplitude, which will be denoted by  $A_{2pw}$ , must be determined. However, the Kirchhoff diffraction theory is valid for a field obeying the linear wave equation, so it cannot be applied directly to the generated second harmonic field.

A field that does allow determination of the second harmonic plane wave amplitude from a measurement over the receiver would be a

plane wave having amplitude  $A_{2pw}$  and frequency equal to the second harmonic frequency, obeying the linear wave equation, passing through an aperture of diameter equal to that of the transducer. For this linear field the measured value over the receiver could be corrected by the Lommel diffraction correction to calculate  $A_{2pw}$ . If it can be shown that the measured value over the receiver of the second harmonic field in the actual physical case is nearly the same as the measured value over the receiver for the linear wave, then the measured value can be corrected by the Lommel diffraction correction to obtain the second harmonic plane wave amplitude  $A_{2pw}$ .

The value measured at the receiver depends on the phase relationships of the second harmonic spherical wavelets and the amplitudes of the wavelets. For both the physical case and the linear wave the individual wavelets are spherical, and the frequencies of the wavelets are the same in the two cases. Therefore the phase relationships are the same.

For the linear wave the amplitude of the spherical wavelets is  $(A_{2pw}/r)$ ,  $r$  being the distance from the source element. In the physical case the amplitude of the wavelets is some function of  $r$ , divided by  $r$ . Since all the spherical wavelets reaching the receiver travel about the same distance, the difference in amplitude between different wavelets due to second harmonic generation is small. Therefore the amplitude of the wavelets, at the position of the receiver, can be approximated by a constant divided by  $r$ . This constant is equal to  $A_{2pw}$ , and the amplitude of the wavelets is  $A_{2pw}/r$ , because that is

the amplitude the spherical wavelets of an infinitely extended plane wave would have in order to produce a plane wave of amplitude  $A_{2pw}$ . Thus the amplitudes of the wavelets for the physical case and the linear wave are nearly the same.

The values of the nonlinearity parameter obtained by correcting for diffraction both the fundamental and second harmonic components are denoted by  $\beta_{c,1,2}$ :

$$\beta_{c,1,2} = \frac{|D_L|_1^2 \beta_u}{|D_L|_2} = \frac{\beta_{c,1}}{|D_L|_2}. \quad (IV.3)$$

The values of  $\beta_{c,1,2}$  are listed in Table IV-2 along with the values of  $s_2$  and  $|D_L|_2$ , the subscript 2 referring to the second harmonic component, and the  $\beta_{c,1,2}$  values are plotted as a function of transducer diameter in Figure IV-10. It is seen from the graph and the last column of Table IV-2, the percent deviation from the mean, that the values are reasonably consistent as a function of transducer diameter, except that the value obtained with the smallest (0.189 cm) diameter transducer is somewhat higher than the others. The mean of these corrected values, 6.159, is about 11% higher than the mean of the values that were corrected for diffraction of the fundamental only. Using this mean value of  $\beta_{c,1,2}$  to calculate  $K_3$ , the result is  $K_3 = (-15.4 \pm 0.8) \times 10^1 \text{ es.cm}^2$ .

The results obtained by correcting for diffraction of only the fundamental component were discussed earlier in this section. Those results were consistent as a function of transducer diameter and produced a value of  $K_3$  which agreed very well with previous, accepted results.

Table IV-2. Values of the Nonlinearity Parameter Corrected for Diffraction of Both the Fundamental and Second Harmonic Components and Quantities Used in the Calculation\*

Transducer Diameter (cm)	$S_2$	$ D_L _2$	$\beta_{c,1,2}$	Percent Deviation From the Mean
1.27	0.0305	0.962	$5.701 \pm 0.036$	7
1.15	0.0374	0.958	$5.853 \pm 0.052$	5
1.01	0.0477	0.952	$6.160 \pm 0.156$	0
0.891	0.0618	0.945	$5.666 \pm 0.073$	8
0.640	0.1171	0.926	$6.066 \pm 0.065$	2
0.512	0.1868	0.907	$6.225 \pm 0.085$	1
0.385	0.3240	0.879	$6.119 \pm 0.043$	1
0.258	0.7217	0.835	$6.334 \pm 0.093$	3
0.189	1.3436	0.764	$7.311 \pm 0.060$	19

\*Each uncertainty given is one standard deviation using (N-1) weighting.

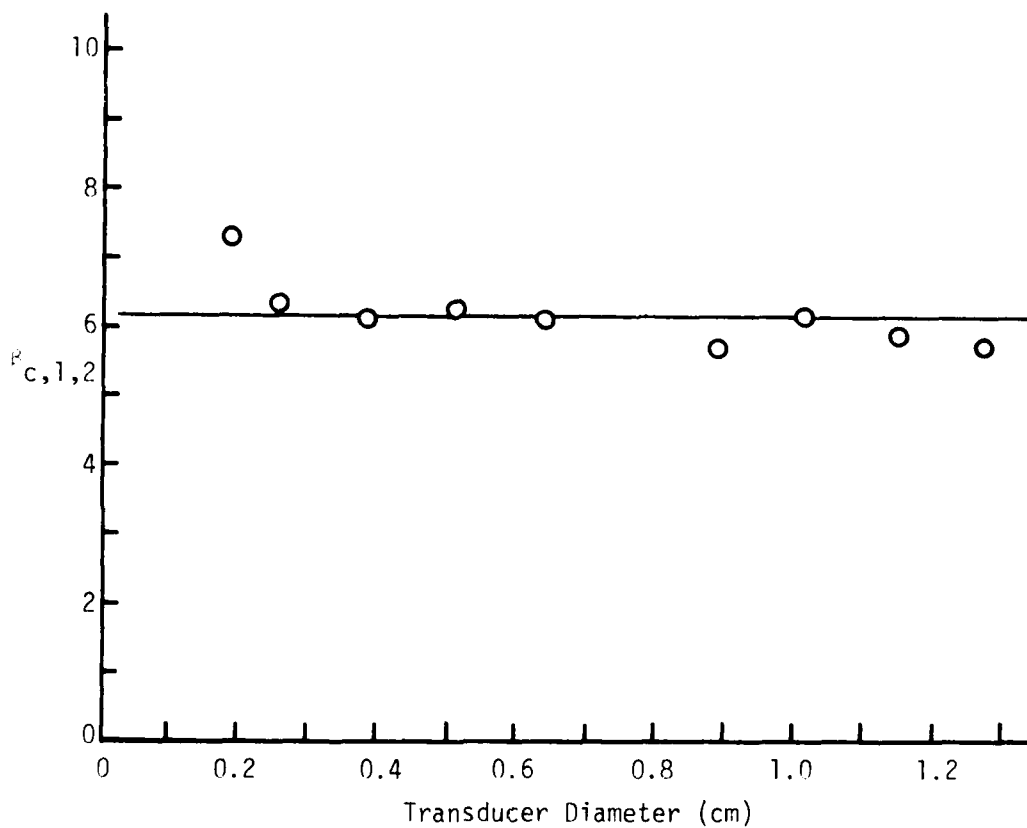


Figure IV-10.  $R_{C,1,2}$  versus transducer diameter for Cu [100]. The straight line represents the mean value  $\bar{R}_{C,1,2} = 6.159$ .

This indicated that a correction obtained by also considering the diffraction of the second harmonic component should be small and the corrected results should be consistent as a function of transducer diameter. The proposed model for the diffraction of the second harmonic gives results which meet both of these desired conditions. The method in which only the fundamental component was corrected is considered to be the more dependable of the two correction methods.

It is seen in Figure IV-10 that, although the  $\beta_{c,1,2}$  values are reasonably consistent as a function of diameter, the values appear to approach the mean value obtained with the previous correction,  $\bar{\beta}_{c,1} = 5.536$ , as the diameter increases. This trend tends to support the use of the correction of only the fundamental, and it indicates that the two correction methods are in good agreement for the larger diameters. The trend suggests that the theory that was used to obtain the  $\beta_{c,1,2}$  values may be slightly less applicable for the smaller diameters than for the larger diameters. Also, since the values of  $\beta_{c,1,2}$  for the smaller diameters are larger than the corresponding values of  $\beta_{c,1}$ , which agreed very well with accepted results, the second correction may be a slight overcorrection.

#### IV. RESULTS OF THE MEASUREMENTS ON $\text{CsCdF}_3$ AND $\text{KZnF}_3$

The samples of  $\text{CsCdF}_3$  and  $\text{KZnF}_3$ , described in Chapter III, are small enough that the data obtained on them require corrections for diffraction. The data were taken and corrected in the same manner as the Cu [100] data. The values of  $A_{1u}$  and  $A_{2u}$ , calculated with no

correction for diffraction, are listed in Table IV-3 along with the quantity  $(8A_{2u}/A_{1u}^2 k^2 a)$ . Plots of  $A_{2u}$  versus  $A_{1u}^2 k^2 a$  are shown in Figures IV-11 through IV-17. After correcting the  $A_{2u}$  values by subtracting the  $A_{2u}$ -intercepts, the nonlinearity parameters were calculated and plotted as a function of  $F_{1u}$  (Figures IV-18 through IV-24). As with the copper data, the uncorrected value of the nonlinearity parameter was taken to be the mean of these calculated values. The data were then corrected for diffraction of the fundamental component and for diffraction of both the fundamental and second harmonic components, as was done for the copper data. The pertinent quantities and results of the calculations for  $\beta$  and  $K_3$  are listed in Table IV-4. The results of the measurements in the [100] and [110] directions in  $\text{CsCdF}_3$  using two different diameter transducers are in good agreement. Notice that the percent differences between the values of  $K_3$  obtained by correcting only the fundamental for diffraction ( $K_{3(1)}$ ) and the values obtained by correcting both the fundamental and second harmonic ( $K_{3(1,2)}$ ) are all less than 9%. In Table IV-5, values of combinations of third-order elastic constants are given, obtained using the correction of the fundamental component only.

The values of  $\beta$  and  $K_3$  presented in Table IV-4, obtained using the diffraction correction method discussed herein, are the values that are being reported as a result of this research. It was indicated in Section IV-1 that a possible approach to determining the nonlinearity parameter of a small sample would be the use of a calibration curve. For comparison with the results given in Table IV-4, the calibration correction is discussed here. The data presented in Figure IV-8 on p. 49 is shown in Figure IV-25 with a smooth curve drawn through the

Table IV-3. Values of  $A_{1u}$ ,  $A_{2u}$ , and  $(8 A_{2u}/A_{1u}^2 k^2 a)$  for  $\text{CsCdF}_3$  and  $\text{KZnF}_3$

Sample, Direction, Transducer Diameter (cm)	Fundamental Amplitude $A_{1u}$ ( $10^{-10}$ m)	Second Harmonic Amplitude $A_{2u}$ ( $10^{-12}$ m)	$\frac{8A_{2u}}{A_{1u}^2 k^2 a}$
$\text{CsCdF}_3$ , [100], 0.512	1.493	0.891	11.998
	1.793	1.249	11.672
	2.029	1.555	11.348
	2.108	1.744	11.790
	2.364	2.215	11.907
	2.580	2.603	11.745
	2.817	3.134	11.863
	2.974	3.511	11.920
	3.152	3.935	11.899
	3.348	4.477	11.992
$\text{CsCdF}_3$ , [100], 0.385	1.883	1.596	13.008
	2.043	1.853	12.838
	2.216	2.188	12.884
	2.376	2.505	12.829
	2.575	2.978	12.978
	2.709	3.324	13.095
	2.882	3.669	12.772
	3.033	4.121	12.952
	3.210	4.626	12.976
	3.370	5.104	12.992
$\text{CsCdF}_3$ , [110], 0.512	1.914	0.838	7.528
	2.109	1.017	7.525
	2.310	1.201	7.407
	2.510	1.405	7.336
	2.651	1.561	7.310
	2.872	1.813	7.234
	3.012	2.041	7.402
	3.153	2.246	7.433
	3.313	2.462	7.377
	3.534	2.774	7.306
$\text{CsCdF}_3$ , [110], 0.385	2.592	1.589	7.481
	2.771	1.783	7.339
	2.906	2.011	7.530
	3.081	2.234	7.442
	3.224	2.465	7.496
	3.403	2.739	7.475
	3.529	2.980	7.566
	3.681	3.222	7.516
	3.847	3.491	7.455
	3.991	3.705	7.355



Table IV-3 (continued)

Sample, Direction, Transducer Diameter (cm)	Fundamental Amplitude $A_{1u}$ ( $10^{-10}$ m)	Second Harmonic Amplitude $A_{2u}$ ( $10^{-12}$ m)	$\frac{8A_{2u}}{A_{1u}^2 k^2 a}$
CsCdF <sub>3</sub> , [111], 0.385	1.715	0.565	16.426
	1.741	0.618	17.433
	1.802	0.650	17.124
	1.857	0.686	17.012
	1.907	0.738	17.343
	1.967	0.752	16.619
	2.013	0.802	16.937
KZnF <sub>3</sub> , [100], 0.512	1.611	0.532	12.553
	1.715	0.610	12.696
	1.833	0.697	12.700
	2.003	0.838	12.786
	2.147	0.955	12.688
	2.344	1.112	12.397
	2.475	1.252	12.529
	2.618	1.393	12.449
	2.749	1.550	12.560
	2.919	1.730	12.432
KZnF <sub>3</sub> , [110], 0.512	1.715	0.363	8.169
	1.839	0.421	8.231
	1.964	0.490	8.415
	2.068	0.538	8.323
	2.203	0.615	8.386
	2.328	0.684	8.363
	2.432	0.747	8.360
	2.546	0.834	8.516
	2.650	0.884	8.331
	2.775	0.964	8.294

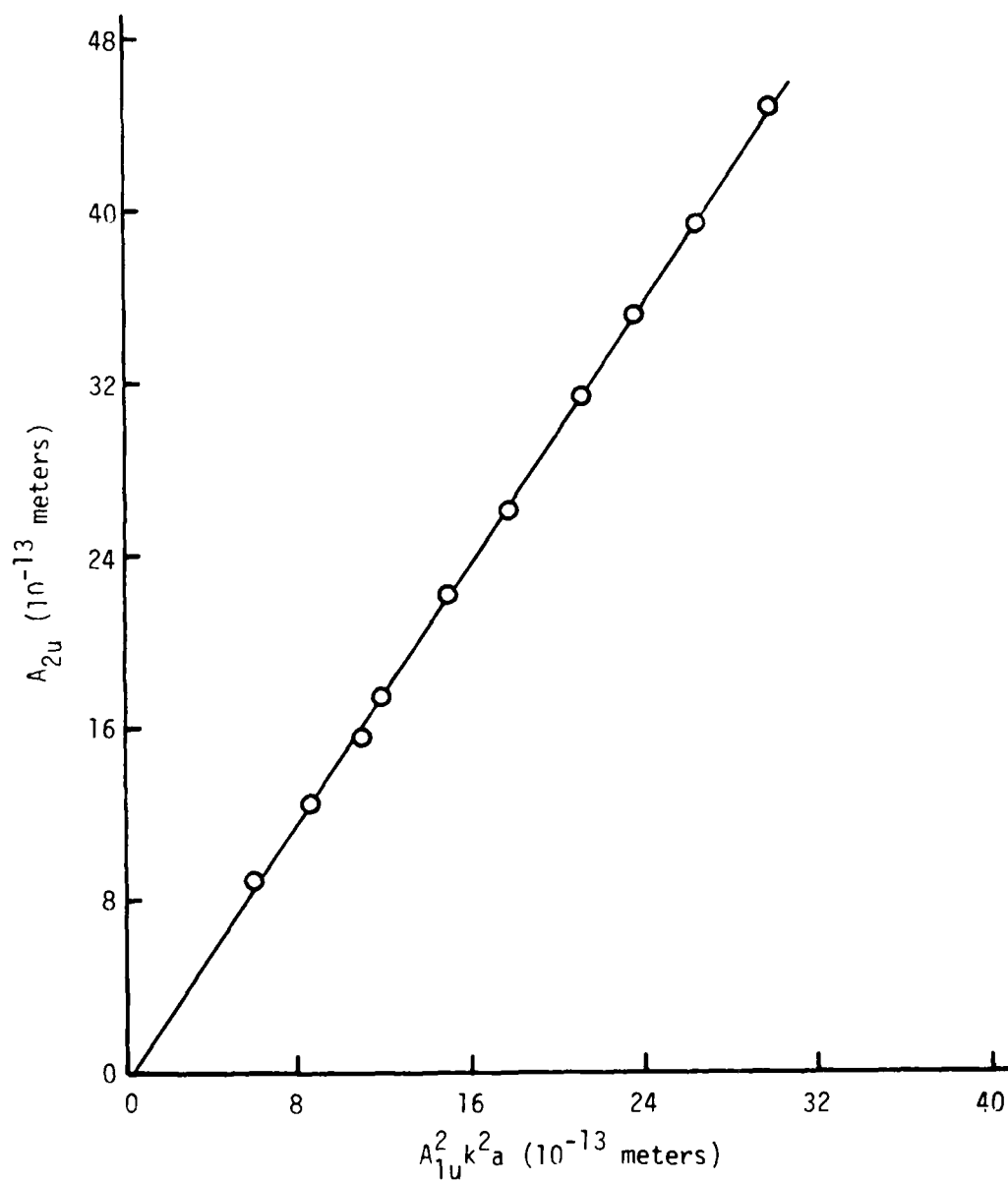


Figure IV-11.  $A_{2u}$  versus  $A_{1u}^2 k^2 a$  for  $\text{CsCdF}_3$  [100] using the 0.51 cm diameter transducer and receiving electrode.

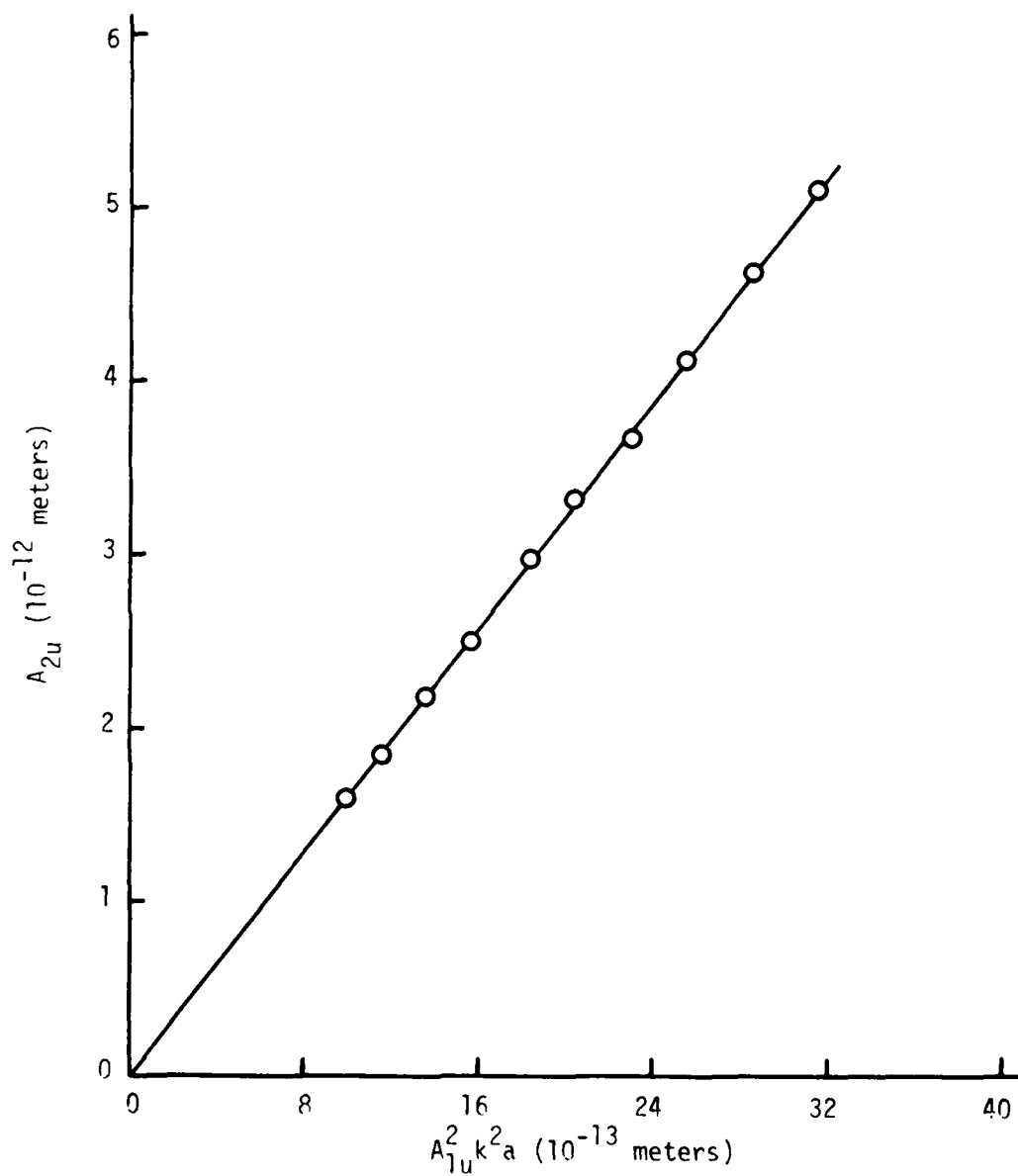


Figure IV-12.  $A_{2u}$  versus  $A_{1u}^2 k^2 a$  for  $\text{CsCdF}_3$  [100] using the 0.38 cm diameter transducer and receiving electrode.

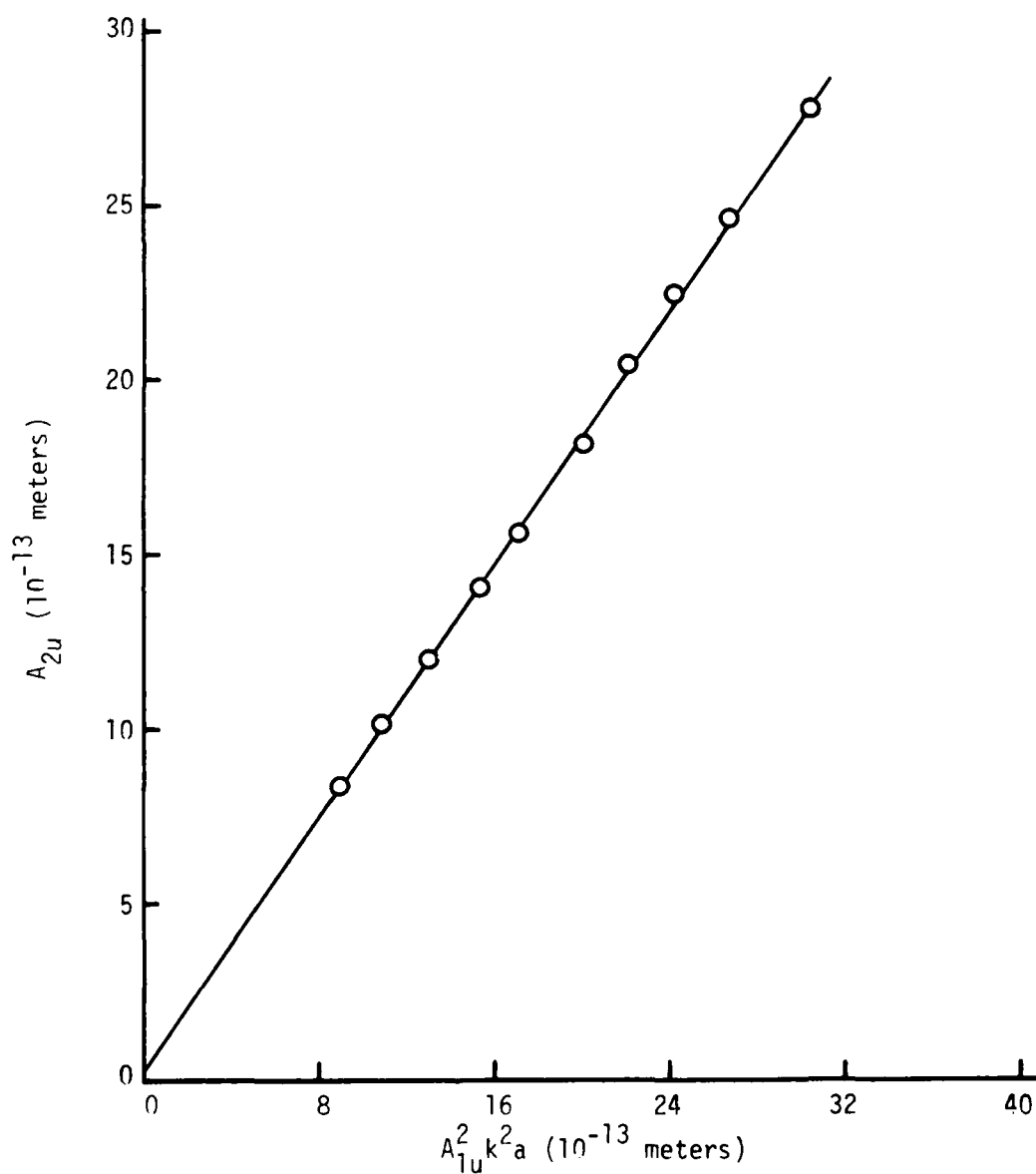


Figure IV-13.  $A_{2u}$  versus  $A_{1u}^2 k^2 a$  for  $\text{CsCdF}_3$  [110] using the 0.51 cm diameter transducer and receiving electrode.

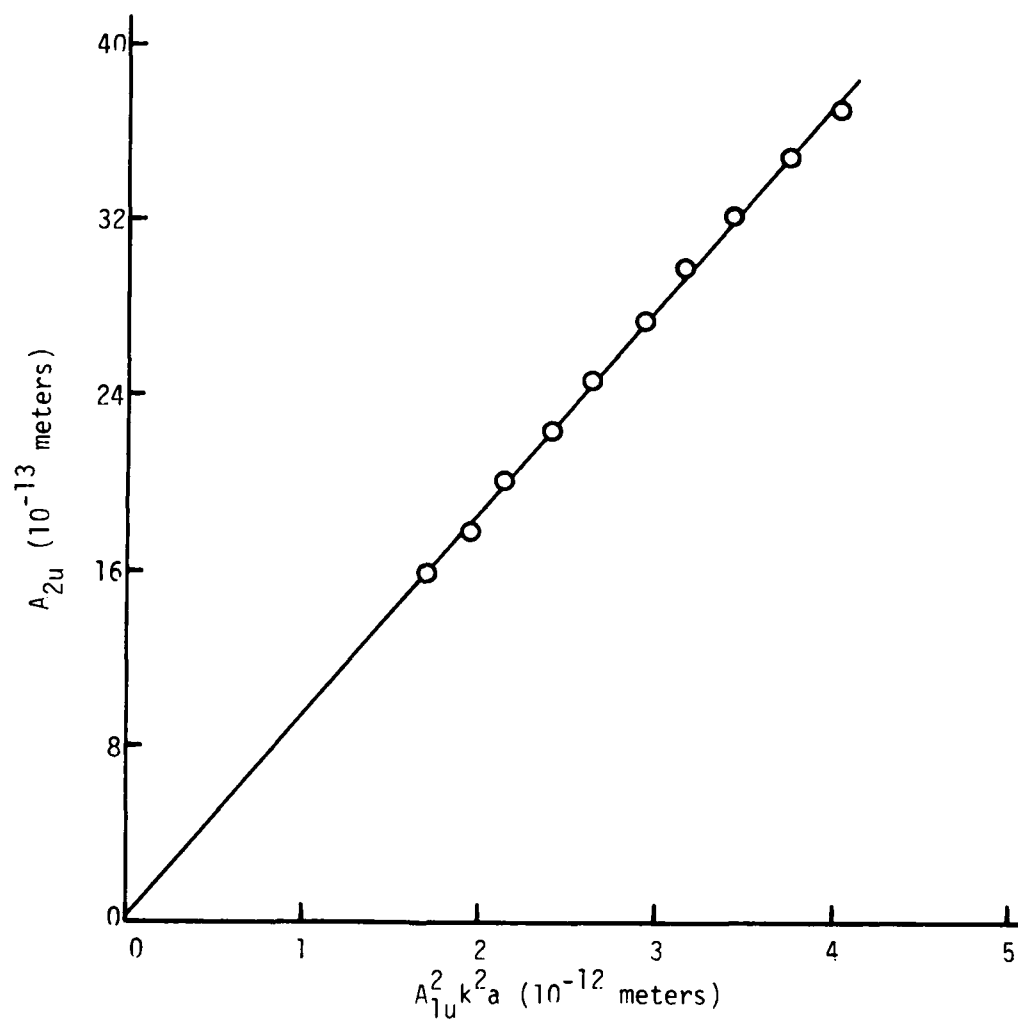


Figure IV-14.  $A_{2u}$  versus  $A_{1u}^2 k^2 a$  for  $\text{CsCdF}_3$  [110] using the 0.38 cm diameter transducer and receiving electrode.

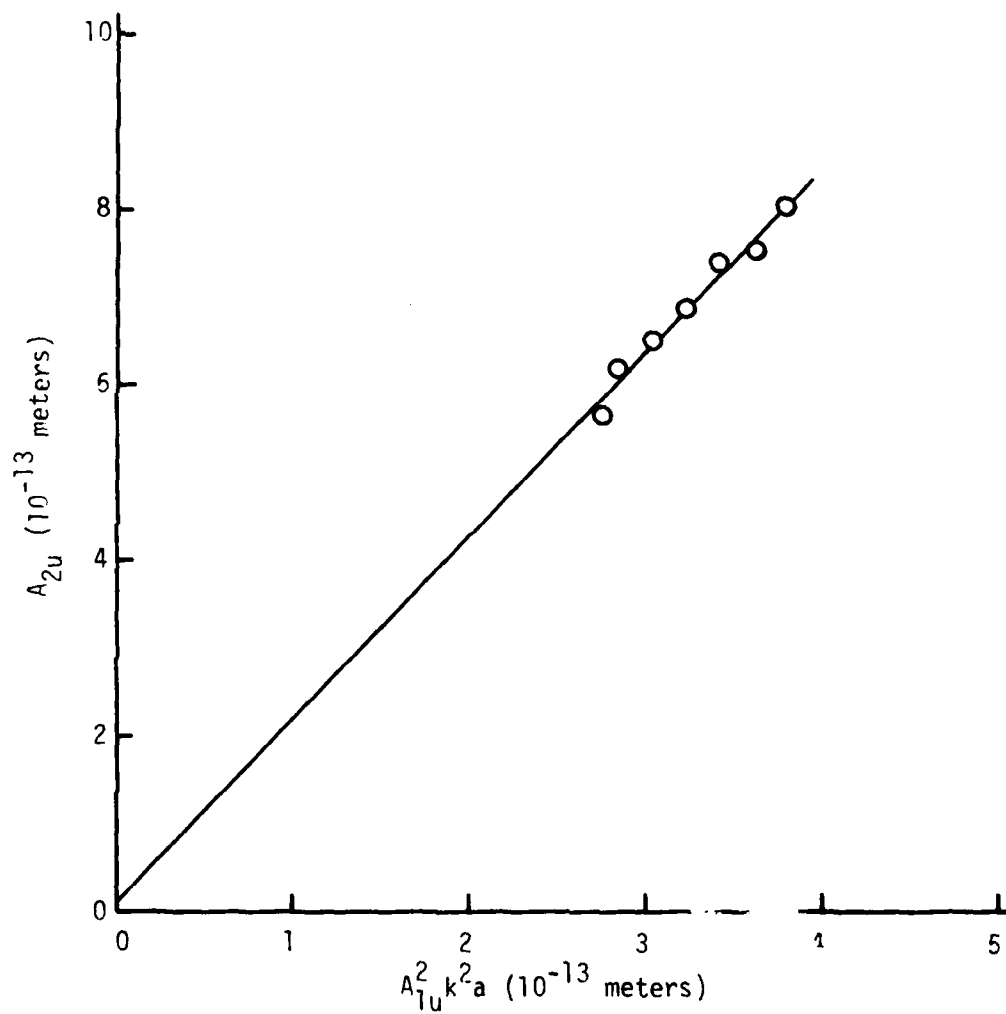


Figure IV-15.  $A_{2u}$  versus  $A_{1u}^2 k^2 a$  for  $\text{CsCdF}_3$  [111] using the 0.38 cm diameter transducer and receiving electrode.

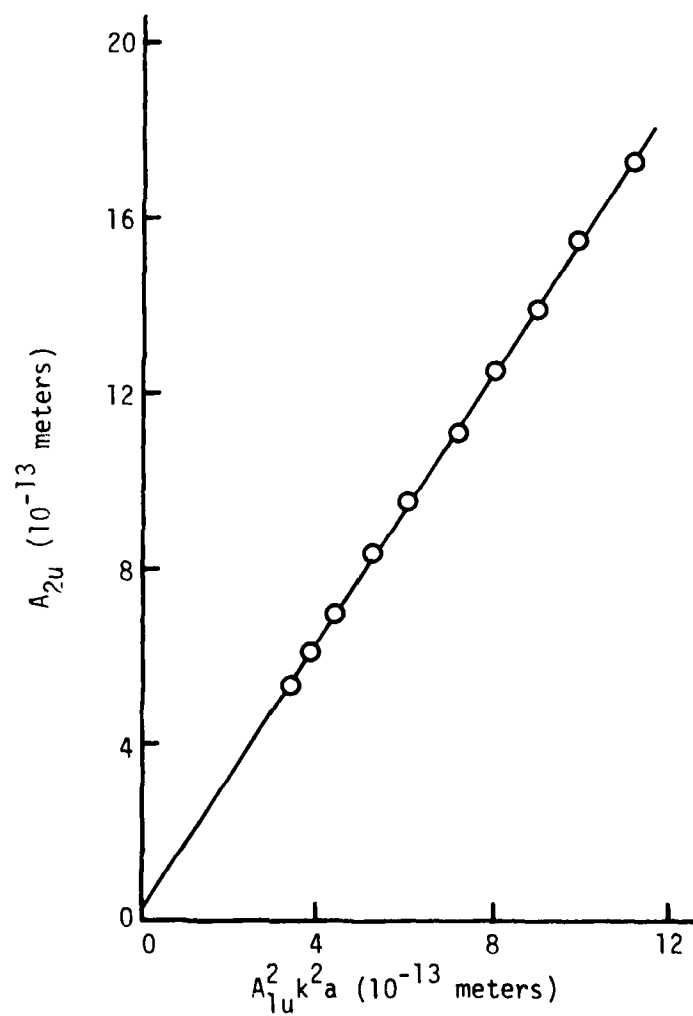


Figure IV-16.  $A_{2u}$  versus  $A_{1u}^2 k^2 a$  for  $\text{KZnF}_3$  [100] using the 0.51 cm diameter transducer and receiving electrode.

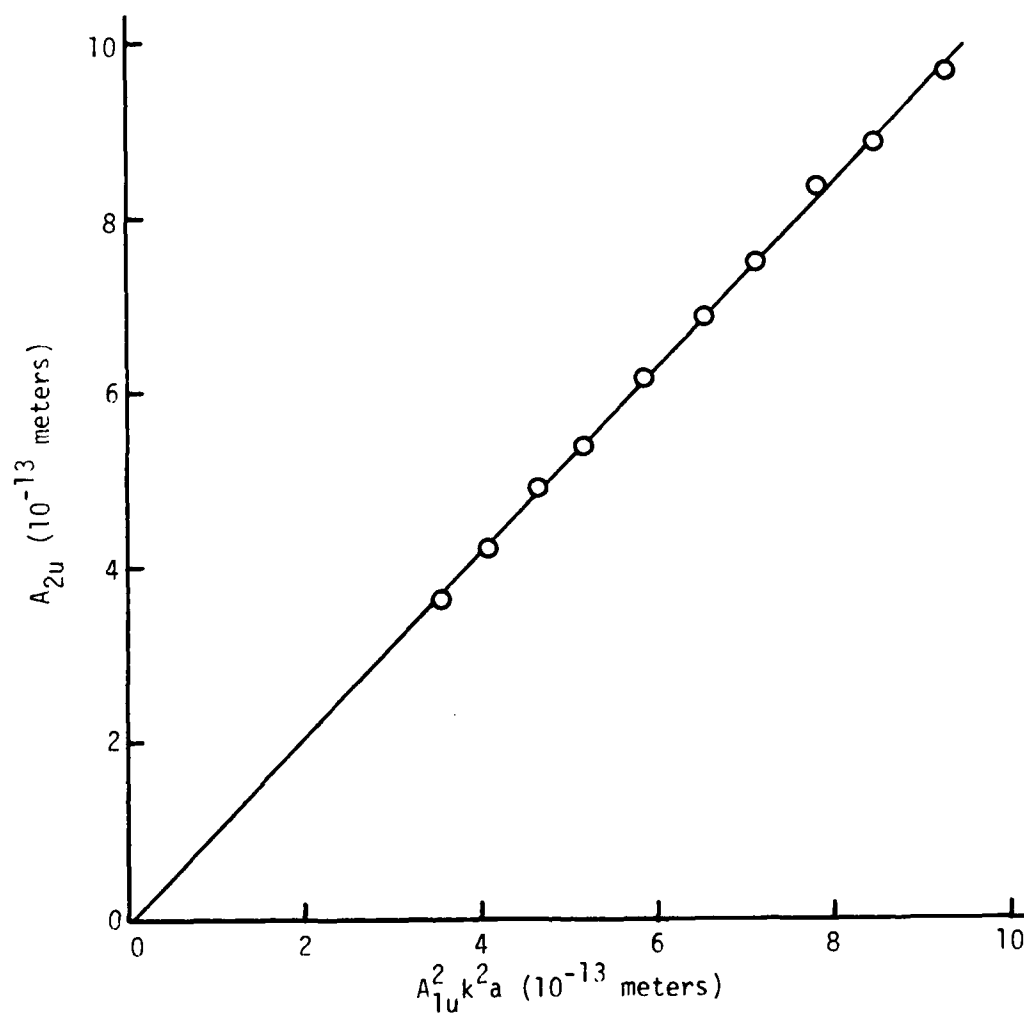


Figure IV-17.  $A_{2u}$  versus  $A_{1u}^2 k^2 a$  for  $KZnF_3$  [110] using the 0.38 cm diameter transducer and receiving electrode.



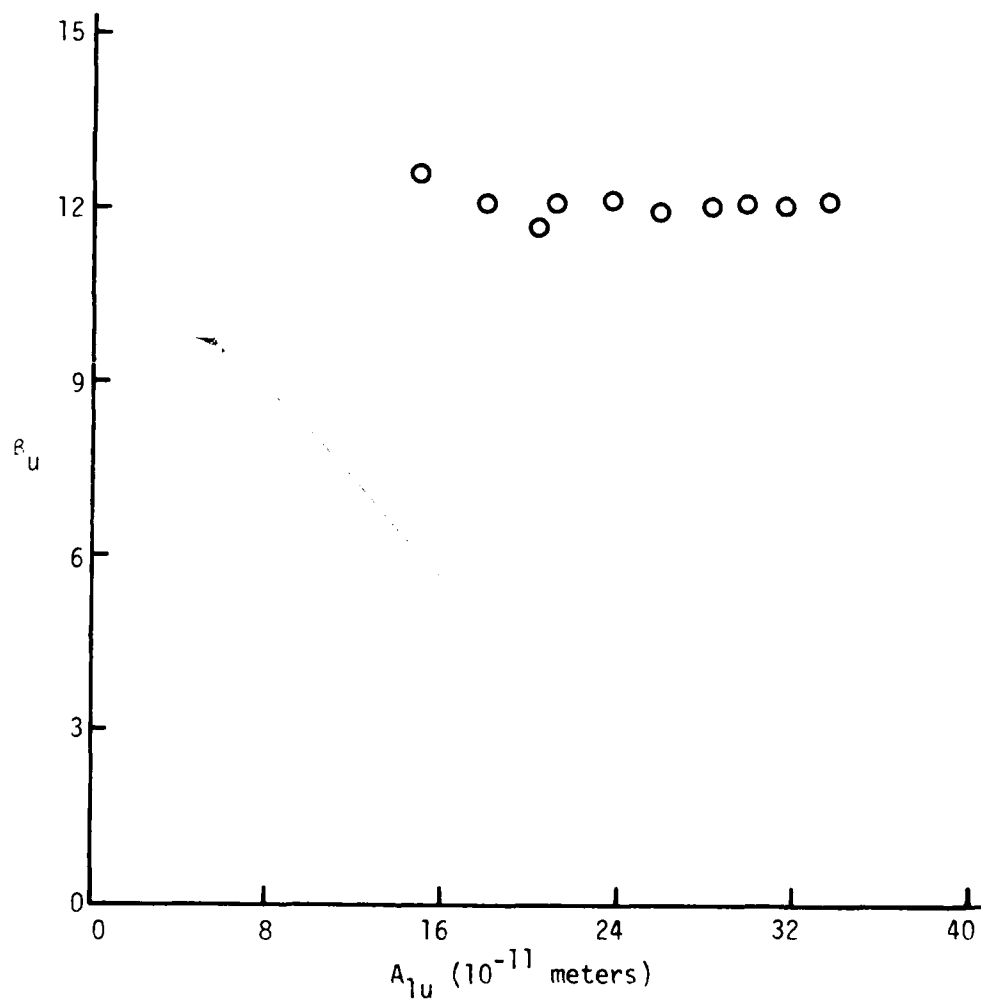


Figure IV-18.  $R_u$  versus  $A_{1u}$  for  $\text{CsCdF}_3$  [100] using the 0.51 cm diameter transducer and receiving electrode.

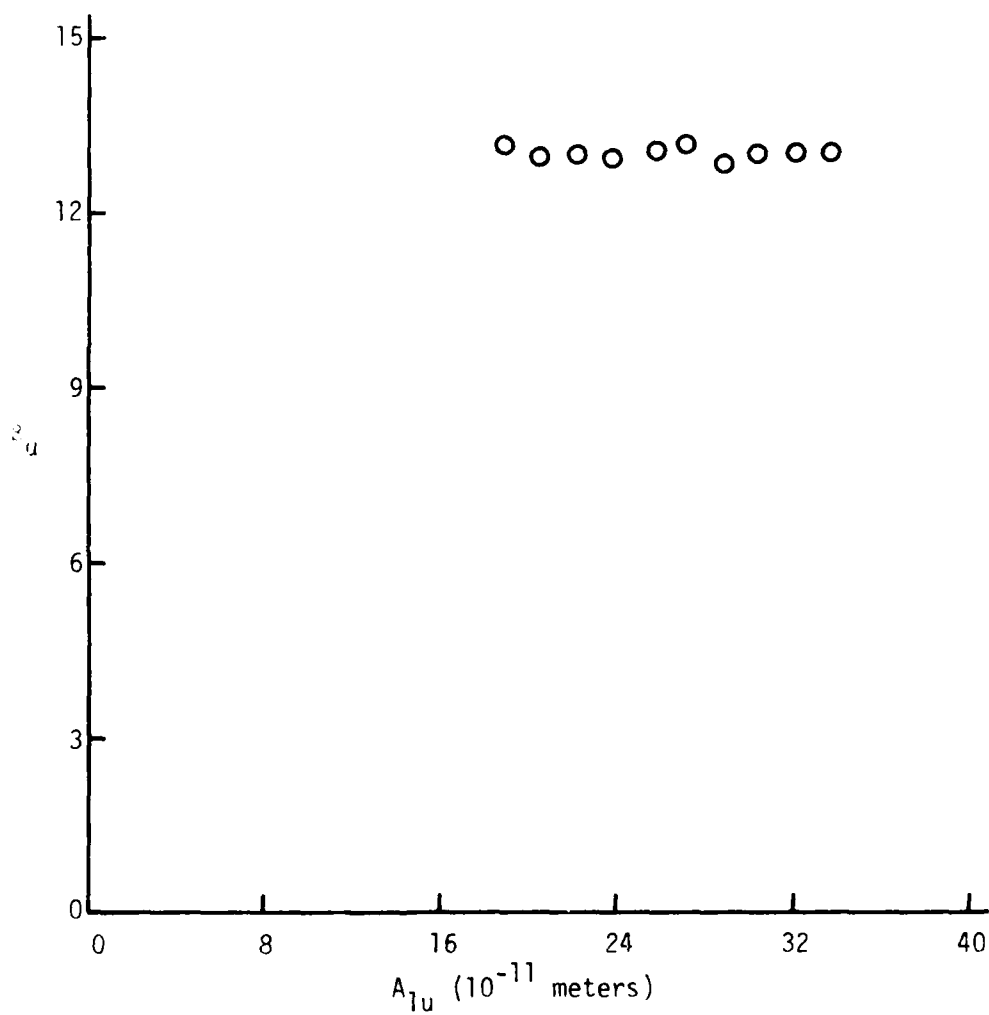


Figure IV-19.  $R_u$  versus  $A_{1u}$  for  $\text{CsCdF}_3$  [100] using the 0.38 cm diameter transducer and receiving electrode.

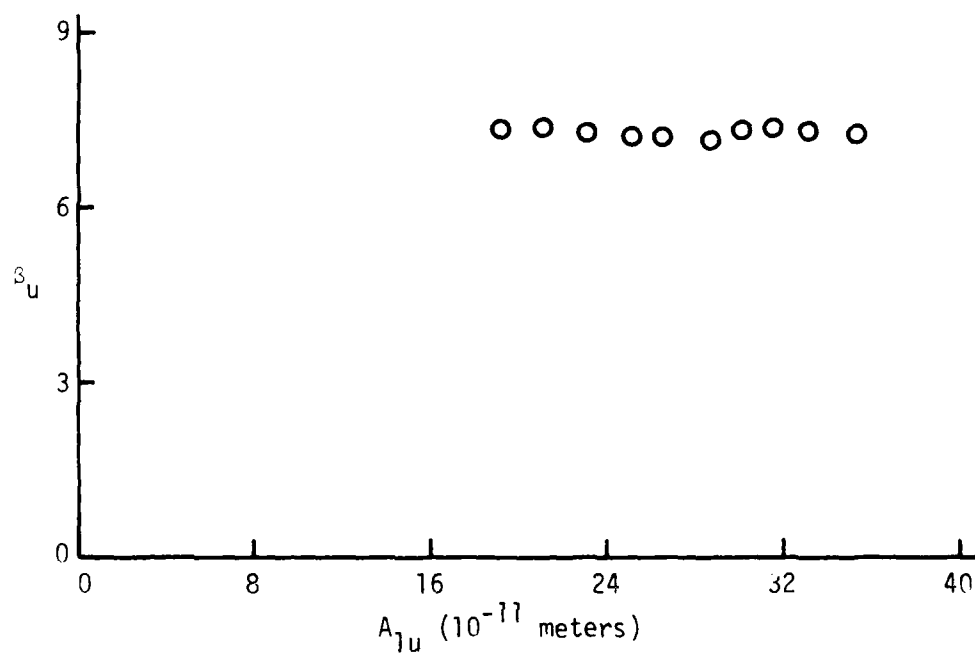


Figure IV-20.  $\beta_u$  versus  $A_{1u}$  for  $\text{CsCdF}_3$  [110] using the 0.51 cm diameter transducer and receiving electrode.

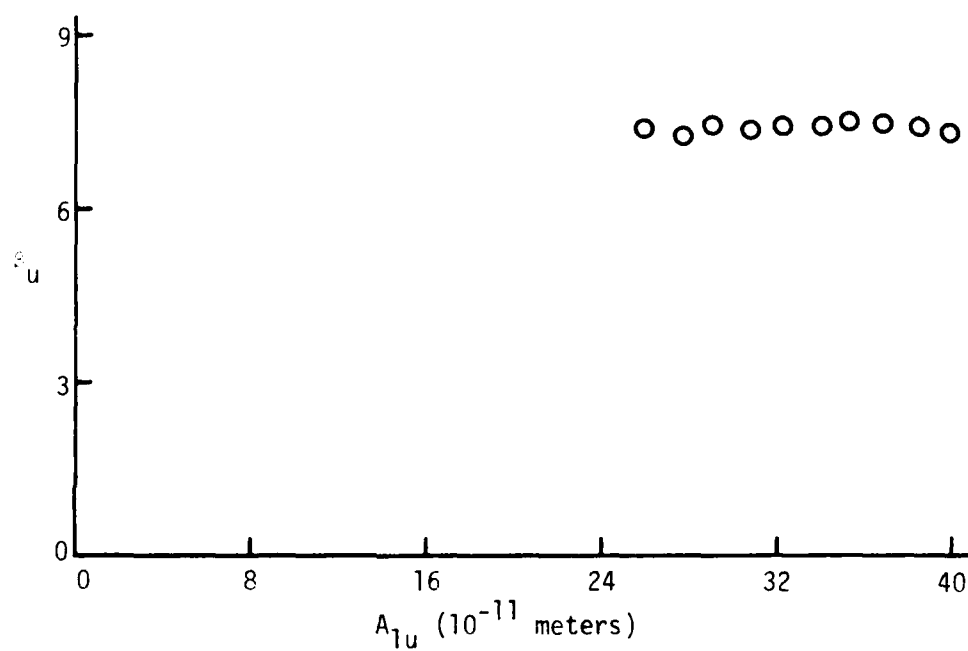


Figure IV-21.  $\beta_u$  versus  $A_{1u}$  for  $\text{CsCdF}_3$  [110] using the 0.38 cm diameter transducer and receiving electrode.

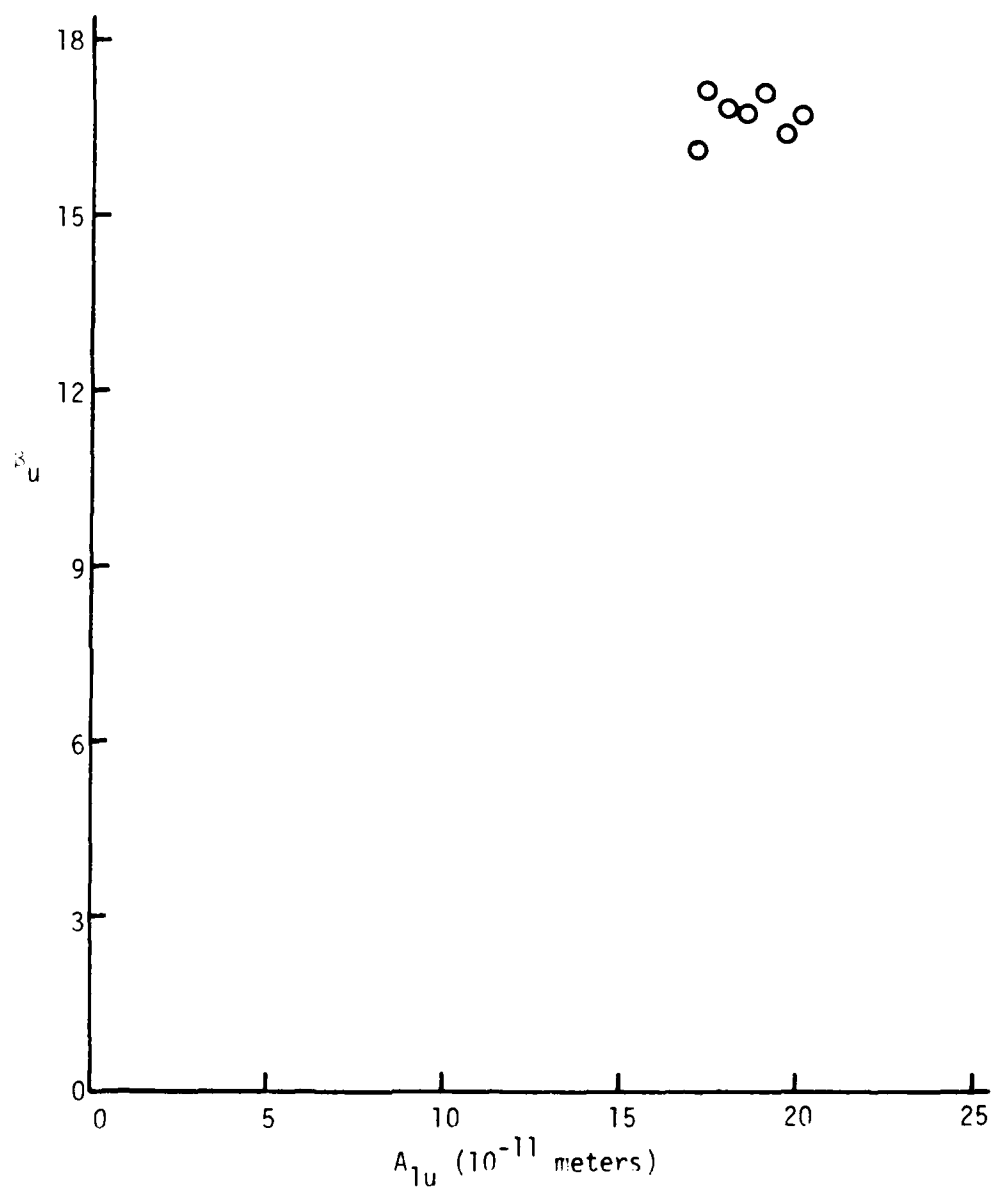


Figure IV-22.  $P_u$  versus  $A_{1u}$  for  $\text{CsCdF}_3$  [111] using the 0.38 cm diameter transducer and receiving electrode.

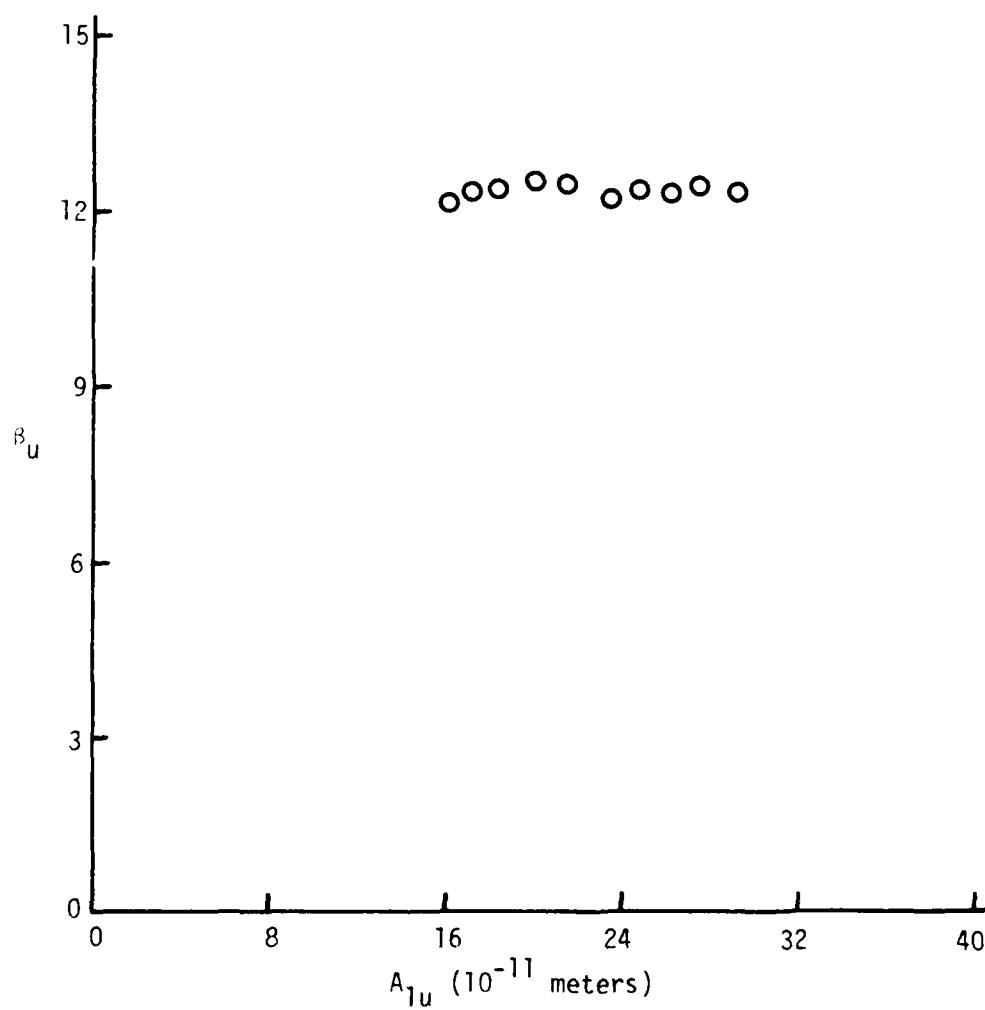


Figure IV-23.  $\beta_u$  versus  $A_{1u}$  for  $\text{KZnF}_3$  [100] using the 0.51 cm diameter transducer and receiving electrode.

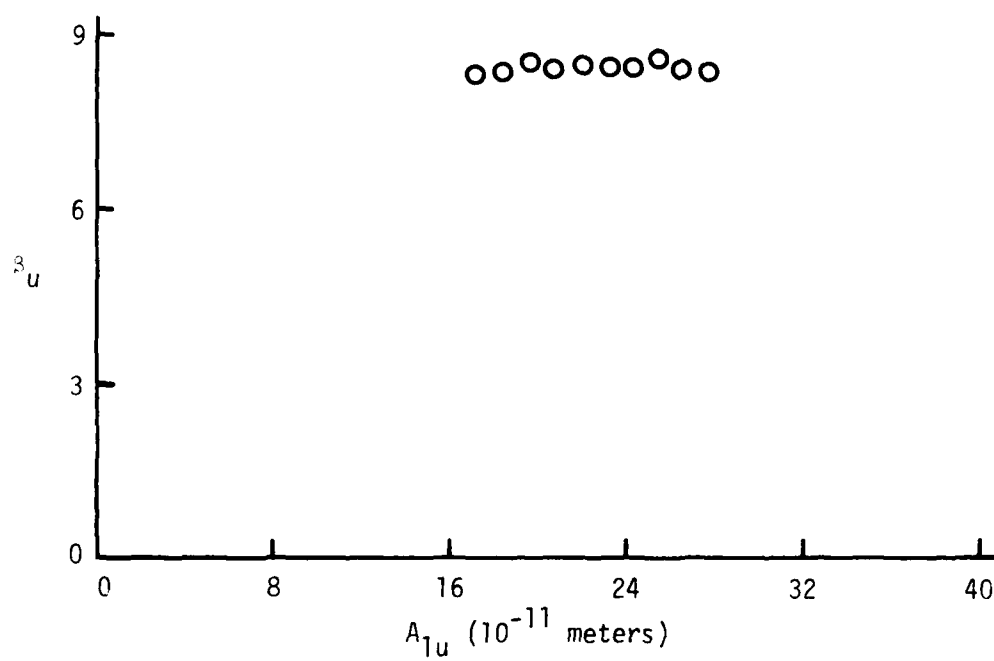


Figure IV-24.  $\beta_u$  versus  $A_{1u}$  for  $\text{KZnF}_3$  [110] using the 0.38 cm diameter transducer and receiving electrode.

Table IV-4. Results of the Measurements on CsCdF<sub>3</sub> and KZnF<sub>3</sub> and Quantities Used in the Calculations\*

Direction	CsCdF <sub>3</sub>					KZnF <sub>3</sub>	
	[100]	[100]	[110]	[110]	[111]	[100]	[110]
Transducer Diameter (cm)	0.512	0.385	0.512	0.385	0.385	0.512	0.512
S <sub>1</sub>	0.3038	0.5271	0.2460	0.4266	0.1504	0.3501	0.2921
S <sub>L1</sub> <sup>1</sup>	0.883	0.855	0.895	0.866	0.916	0.878	0.887
S <sub>L2</sub> <sup>2</sup>	0.781	0.730	0.800	0.750	0.839	0.771	0.787
S <sub>2</sub>	0.1519	0.2635	0.1230	0.2133	0.0752	0.1750	0.1461
S <sub>L12</sub> <sup>12</sup>	0.916	0.891	0.924	0.901	0.940	0.910	0.917
E <sub>0</sub>	12.1 ± 0.22	13.0 ± 0.10	7.28 ± 0.07	7.41 ± 0.07	17.0 ± 0.37	12.3 ± 0.11	8.42 ± 0.08
c <sub>11</sub> <sup>1</sup>	9.43 ± 0.18	9.50 ± 0.07	5.83 ± 0.06	5.56 ± 0.06	14.3 ± 0.31	9.51 ± 0.09	6.63 ± 0.07
c <sub>11,2</sub> <sup>1,2</sup>	10.3 ± 0.19	10.7 ± 0.08	6.31 ± 0.06	6.17 ± 0.06	15.2 ± 0.33	10.4 ± 0.10	7.23 ± 0.07
K <sub>3(1)</sub>	-13.4 ± 0.19	-13.5 ± 0.08	-8.78 ± 0.06	-8.52 ± 0.06	-16.7 ± 0.30	-16.8 ± 0.12	-12.7 ± 0.09
(x10 <sup>12</sup> dyn/cm <sup>2</sup> )							
K <sub>3(1,2)</sub>	-14.4 ± 0.21	-14.8 ± 0.09	-9.26 ± 0.06	-9.13 ± 0.06	-17.6 ± 0.32	-18.1 ± 0.13	-13.5 ± 0.09
(x10 <sup>12</sup> dyn/cm <sup>2</sup> )							
Percent Difference in K <sub>3(1)</sub> and K <sub>3(1,2)</sub>	7	9	5	7	5	7	6

\*Each uncertainty given is one standard deviation using (N-1) weighting.



Table IV-5. Measured Values of Combinations of Third-Order Elastic Constants of  $\text{CsCdF}_3$  and  $\text{KZnF}_3$  in Units of  $10^{12}$  dyn/cm<sup>2</sup>

Sample	$C_{111}$	$(C_{112} + 4C_{166})$	$(6C_{144} + C_{123} + 8C_{456})$
$\text{CsCdF}_3$	-13.4	- 7.24	-46.6
$\text{KZnF}_3$	-16.8	-11.3	

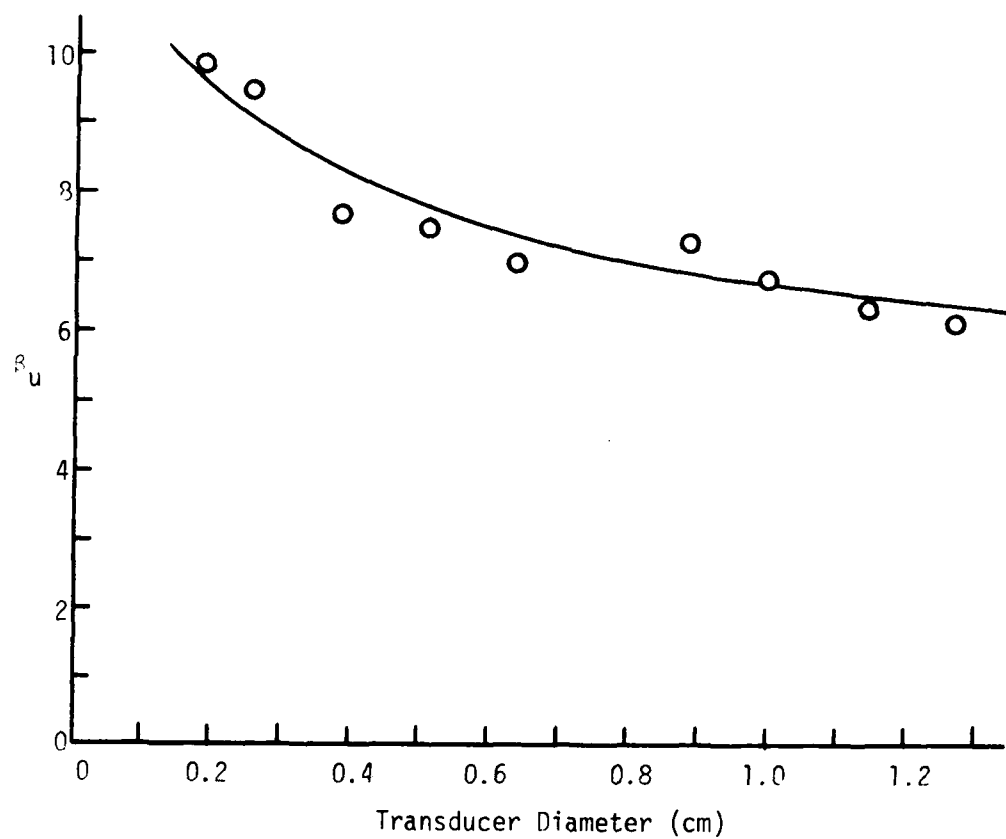


Figure IV-25.  $R_u$  versus transducer diameter for Cu [100]. The smooth curve can be used as a calibration curve.

data points. The data approach a value of the nonlinearity parameter of approximately 5.5. The smooth curve can be used to approximate a correction factor for data obtained using a transducer-receiver pair of given diameter. The values on the smooth curve at 0.385 cm and 0.512 cm, the diameters used for the measurements on  $\text{CsCdF}_3$  and  $\text{KZnF}_3$ , are  $\sim 8.3$  and  $\sim 7.8$ . The correction factor would be  $5.5/8.3 = .66$  for the 0.385 cm diameter and  $5.5/7.8 = .71$  for the 0.512 cm diameter. The correction factors obtained from the Lommel diffraction correction,  $|D_L|_1^2$  in Table IV-4, range from 0.730 to 0.839 for the 0.385 cm diameter and from 0.771 to 0.800 for the 0.512 cm diameter. The values of  $K_3$  calculated from the calibration correction factors, listed in the same order as that in Table IV-4, are -12.5, -12.5, -8.13, -7.85, -13.7, -15.8, and -11.8, in units of  $10^{12}$  dynes/cm<sup>2</sup>. The correction factors obtained from the calibration curve do not take into account the length of the sample or the wavelength of a given frequency wave in the material. These quantities are taken into account by the Lommel diffraction correction.

#### V. ESTIMATE OF THE DIMENSIONS OF THE SMALLEST MEASURABLE SAMPLE

It is useful to estimate the dimensions of the smallest sample measurable by the present technique. The minimum dimension transverse to the direction of wave propagation is approximately 5 mm; this allows the sample walls to be sufficiently outside the ultrasonic beam and is large enough for measurement with the 0.19 cm transducer and receiver. Equation (II.15) shows that the amplitude of the second harmonic

component in a plane wave is directly proportional to the length of the sample and depends on the quantities  $K_2$  and  $K_3$ ; thus the sample must be long enough to allow a measurable second harmonic to be generated, and this minimum length will depend on the material being measured. It is estimated that this minimum length would be typically of the order of 4 mm.

It may not be possible to perform the measurements on a sample having both the minimum transverse dimension and minimum length given above. Measurement of smaller amplitudes tends to require larger diameter transducer-receiver sets. For example, the [111]  $\text{CsCdF}_3$  sample of length 4.19 mm was measured with the 0.38 cm diameter transducer and receiver, but data could not be obtained using the 0.19 cm or the 0.26 cm diameter transducer and receiver.

## VI. ERROR ANALYSIS

The primary sources of systematic error are the measurement of the substitutional signal voltage and the measurement of the impedance of the resistor in the base of the capacitive detector housing as a function of frequency. It is estimated that the systematic error for the measurement of  $\beta_u$  is within  $\pm 12\%$ . After correction of  $\beta_u$  for diffraction, the mean value  $\bar{\beta}_{c,1}$  agreed very well with previous, accepted results (see Section IV-III), and the largest deviation of any of the nine  $\beta_{c,1}$  values from  $\bar{\beta}_{c,1}$  was 6% (see Table IV-1, p. 53). Therefore the error introduced into the determination of the nonlinearity parameter by the diffraction correction appears to be relatively small,

since the error of the  $\beta_{c,1}$  values was well within the  $\pm 12\%$  systematic error in each case.

#### VII. SOME PROBLEMS ASSOCIATED WITH THE APPLICATION OF DIFFRACTION THEORY TO THE PRESENT MEASUREMENTS

The nonlinear wave equation (II.13) that has been used to describe the wave propagation is valid only for a pure longitudinal mode plane wave. It was obtained from the general equation, (II.12), by assuming that the only component of displacement, say  $u_1$ , is along the pure mode direction, so that  $u_2 = u_3 = 0$ , and that  $u_1$  is a function only of  $a_1$  and  $t$ . However, in the diffraction problem the displacement also includes the components  $u_2$  and  $u_3$ , and each displacement component is a function of  $a_1, a_2, a_3$ , and  $t$ . Thus Eq. (II.12) contains many terms in the general case which are not present when considering only pure longitudinal plane waves. From Eq. (II.12) three long coupled equations are obtained, one for each of the directions labelled 1, 2, 3 [see Gauster (1966)]. An attempt to solve the general equations would be complicated, and would involve nonpure mode propagation.

The previously discussed theory that was used for the diffraction correction was developed assuming the medium to be a homogeneous fluid. The measurements of third-order elastic constants are performed on solid media. At the surface of the solid a measurement is made of displacement, a vector quantity. The measurement technique is sensitive only to the component of displacement normal to the surface. Thus when the transducers are large enough that the waves can be considered plane, the measured value accurately represents the displacement field, but

when the waves can no longer be considered plane, only the longitudinal component is measured. In contrast, the parameter measured for a fluid medium would be a scalar such as pressure or velocity potential. Thus the fluid theory treats the measurement of a scalar quantity instead of a vector quantity.

The anisotropy of the solid also leads to problems. The wave speed is a function of direction in an anisotropic material, and therefore the assumption of spherical Huyghens' wavelets is not strictly valid. Also, since parts of the diffracted wave travel in nonpure mode directions, there is coupling to transverse modes.

Despite the approximations that must be made to treat the effects of diffraction on the nonlinearity measurements, it has been demonstrated that meaningful measurements can be made using the technique presented. The data obtained in the present experiments are presented as a contribution to the solution of the general problem of the diffraction of waves propagating in a nonlinear medium.

#### VIII. SUGGESTIONS FOR FURTHER WORK

A result of the present research was to extend the harmonic generation technique for nonlinearity measurements to include measurement of smaller samples than could be measured previously. Consequently, future measurements can include a wider range of materials. The previously existing harmonic generation technique is capable of performing the nonlinearity measurements as a function of temperature down to liquid helium temperature. The temperature dependent measurements can be extended to include small samples by modifying part of the

cryogenic apparatus and utilizing the diffraction correction. This work has been begun by Jacob Philip.

Measurements in the [100] and [110] directions in  $\text{KZnF}_3$  are reported here. To obtain a third combination of third-order elastic constants a measurement in the [111] direction is needed.

A more rigorous mathematical treatment of the diffraction of finite amplitude waves would be desirable. A tutorial paper by Bajak and Breazeale (1980) presents a quantum mechanical approach to the same problem of plane wave harmonic generation that was treated classically in Chapter II. In the quantum mechanical theory, two phonons of the fundamental frequency interact to produce a phonon of the second harmonic frequency. In the paper there is given an expression for the amplitude of the second harmonic component as a function of the deviation of the direction of the second harmonic wave vector from the direction of the wave vector composed of the sum of the two fundamental frequency wave vectors. This suggests the possibility to correct for diffraction of the second harmonic. A means of implementing the correction for the present experimental configuration would involve resolving the fundamental field into an equivalent set of plane waves and determining the amplitudes of the phonons generated by the many interactions of the phonons in the fundamental field.

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APPENDIX

# APPENDIX

## COMPUTER PROGRAM FOR THE CALCULATION OF THE MAGNITUDE OF THE DIFFRACTION CORRECTION, $|D_L|$ , AS A FUNCTION OF S

The following computer program, DIFCOR.F10, and subroutine, BES.F10, written in FORTRAN for use on the DECsystem-10 Operating System, is used to calculate the magnitude of the diffraction correction,  $|D_L|$ , as expressed in Eq. (IV.1), as a function of s. As written, the value of s varies from 0.00 to 74.99 in increments of 0.01. The program is adapted from that written by D. W. Fitting.

```

00100      REAL J0,J1
00200      DIMENSION DIFCOR(7500),RINDEX(750)
00300      DATA PI/3.1415926/
00400      OPEN(UNIT=25,FILE='DIFCOR.OUT',ACCESS='SEQUOU
T',
00500          * DEVICE='DSKB:')
00600      DO 10 I=1,7499
00700          S=FLOAT(I)/100.
00800          ARG=2.*PI/S
00900          CALL BES(ARG,J0,J1)
01000      10 DIFCOR(I)= (SQRT((COS(ARG)-J0)**2+(SIN(ARG)-J1
)**2))
01100          DIFCOR(0)=1.0
01200      DO 20 I=1,750
01300      20 RINDEX(I)=FLOAT(I-1)/10.
01400          TYPE 6,(RINDEX(I),(DIFCOR(K),K=(I-1)*10,(I-1
)*10+9),
01500          * I=1,750)
01600      6 FORMAT(1H1/1H0,36X,'DIFFRACTION CORRECTION DL
AS A FUNCTION OF S
01700          * '/1H0,3X,'S',7X,'0.00',6X,'0.01',6X,'0.02',
01800          1 6X,'0.03',6X,'0.04',6X,'0.05',6X,'0.06',6X,
01900          2 '0.07',6X,'0.08',6X,'0.09'/1H0/50(3X,F4.1,10F
10.3/))
02000      END

```

```

00100      SUBROUTINE BES(X,J0,J1)
00200      DIMENSION A(7),B(7),C(7),D(7),E(7),F(7)
00300      REAL J0,J1
00400      DATA A/1.0,-.25,1.562495E-2,-4.34008E-4,6.77
4562E-6,
00500      1 -6.679876E-8,3.95152E-10/,B/.5,-6.249998E-2,2
.604145E-3,
00600      2 -5.424265E-5,6.756882E-7,-5.378753E-9,2.08677
9E-11/,
00700      3 C/7.978846E-1,-2.31E-6,-4.97466E-2,-2.56824E-
3,1.11162E-1,
00800      4 -1.769162E-1,1.0553E-1/,D/-7.853982E-1,-1.249
919E-1,
00900      5 -3.5586E-4,7.089471E-2,-4.384125E-2,-7.127919
E-2,
01000      6 9.883782E-2/,E/7.978846E-1,4.68E-6,1.4937E-1,
4.61835E-3,
01100      7 -2.021039E-1,2.761768E-1,-1.460406E-1/,F/-2.3
56194,
01200      8 3.749884E-1,5.085E-4,-1.722273E-1,6.022188E-2
,1.939723E-1,
01300      9 -2.126201E-1/
01400      IF(X.GT.3.0) GO TO 10
01500      J0=A(1)+X*X*(A(2)+X*X*(A(3)+X*X*(A(4)+X*X*(A
(5)+X*X*(A(6)+
01600      * X*X*A(7))))))
01700      J1=X*(B(1)+X*X*(B(2)+X*X*(B(3)+X*X*(B(4)+X*X
*(B(5)+X*X*(B(6)+
01800      * X*X*B(7))))))
01900      RETURN
02000      10 F0=((((C(7)/X+C(6))/X+C(5))/X+C(4))/X+C(3))/X
+C(2))/X+C(1)
02100      TH0=((((D(7)/X+D(6))/X+D(5))/X+D(4))/X+D(3)
)/X+D(2))/X+
02200      * D(1)+X
02300      F1=((((E(7)/X+E(6))/X+E(5))/X+E(4))/X+E(3))
/X+E(2))/X+E(1)
02400      TH1=((((F(7)/X+F(6))/X+F(5))/X+F(4))/X+F(3)
)/X+F(2))/X+
02500      * F(1)+X
02600      J0=F0*COS(TH0)/SQRT(X)
02700      J1=F1*COS(TH1)/SQRT(X)
02800      RETURN
02900      END

```

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